

**REMARKS**

Applicants' representatives acknowledge with appreciation the communication from Examiner Duc Truong on July 21 and 23 requesting a paper copy of Senior Party Zook (PRC) Motion 2 ((Miscellaneous Motion Requesting Acceptance of an Unintentionally Delayed Request for Priority Benefit), the opportunity to inform Examiner Duc Truong of the duty imposed during Interference 105,555 (MPT) to address allegations raised by Junior Party Zook (PBT) that certain claims lack written description and/or are based upon new matter, and authorization to provide the requested paper copy of PRC's Motion 2 with the paper addressing the outstanding issues raised by the allegations of the Junior Party.

New claims 44-47 have been added. Claims 35 and 43 have been amended. The new claims are all dependent claims and the amended claims have been narrowed by the present amendment. Thus, neither the new claims nor the amended claims raise any new issues of patentability; moreover, the present amendments do not add new matter. The new claims find support in the application as originally filed. For example, support for: claim 44 can be found at page 21, lines 28-29; claim 45 can be found at page 20, line 9; claim 46 can be found at p. 19, line 11-16, and page 20, lines 3-6 and 11-13; and claim 47 can be found at page 34, line 7, to page 35, line 17 (see also page 19, lines 19-23). Similarly, the amendment of previously pending claims 35 and 43 find support at page 20, lines 11-13. Since all of these inventions are reasonably conveyed by the specification, there is no issue of new matter.

**Motion with Petition to Correct Unintentionally Delayed Request for Priority Benefit**

Senior Party Zook (PRC) Motion 2 ((Miscellaneous Motion Requesting Acceptance of an Unintentionally Delayed Request for Priority Benefit) was originally filed November 30, 2007. As requested by the Office, a copy of the previously filed motion and associated papers is

provided herewith in paper form. Applicants submit that the motion and associated papers support the grant of the benefit of priority as set forth therein. Applicants request that the Examiner forward the motion and associated papers to the Petitions branch for processing.

Unfounded Allegations of a Lack of Written Description and/or New Matter

Junior Party Zook (PBT) included in its Preliminary Motion List (Attachment A), dated August 22, 2007, allegations that claim 22 and claims dependent therefrom were unpatentable because “there is no support for the term ‘m’ to be defined as an ‘integer’.” Specifically, PBT alleged that:

claims 22-27 and 35-43 of ... Application 10/302,177 [*sic*], that correspond to the count, are unpatentable for new matter ... The Senior Party’s specification, and the claims as originally filed, recite that “m” is a rational number ... claim 22 was amended to state that “m” “an integer between 0 and 10”. It is axiomatic that “0” is an integer, but not a rational number. [and that] claims 22-27 and 35-43 ... are unpatentable for lack of written description support ... [as] there is no support for the term “m” to be defined as an “integer”.

(Junior Party Zook’s Preliminary Motion List, page 2, line 16, to p. 3, line 8)(emphasis in original) The Board mandated that “PRC sh[ould], upon resumption of ex parte prosecution, inform the Examiner of PBT’s rational number allegation and/or amend the claims to correct the alleged problem.” (Attachment B: Interference 105,555 (MPT), Paper 22, page 3, lines 17-19)

In response, Applicants submit that both the arguments raised by Junior Party Zook stem from a single, fundamental error. Specifically, the error that zero (0) is not a rational number. Various defined, a rational number is “[a] number capable of being expressed as an integer or quotient of integers” (Webster’s II New College Dictionary (3rd Ed. (2005)) or “a number that can be expressed as an integer or the quotient of an integer divided by a nonzero integer” (Merriam-Webster Online Dictionary). Further, an integer is “[a] member of the set of positive

numbers {1, 2, 3, ...}, negative whole numbers {-1, -2, -3, ...}, and zero {0} ..." (Webster's II New College Dictionary (3rd Ed. (2005)) As zero (0) is an integer, by definition, and a rational number is one that can be expressed as the quotient of an integer divided a nonzero integer, again by definition; zero, which can be expressed as zero divided by a nonzero integer, is a rational number.

Stemming from the underlying, fundamental error that zero (0) is not a rational number, the allegations that claims 22-27 and 35-43 are unpatentable for new matter and/or for lack of written description presented during Interference 105,555 (MPT) are unfounded as the phrase "rational number" characterizing the variable "m" in the chemical formula indicates integers, including zero. Applicants request allowance of the claims to issue.

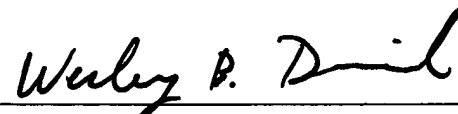
### CONCLUSION

Should this amendment or any of the papers submitted herewith raise any questions, the Examiner is urged to contact Applicants' attorney undersigned below for a telephonic interview to resolve the questions. Please grant any extensions of time required to enter this response and charge any additional required fees to Deposit Account No. 06-0916.

Respectfully submitted,

Date: October 8, 2008

By: \_\_\_\_\_



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Paper No. \_\_\_\_\_

UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

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JONATHAN D. **ZOOK**, DAVID W. JORDAN, DEAN M. WILLARD,  
GEORGE JONES, AND MICHAEL COSMAN,  
Junior Party ("PBT")  
(U.S. Patents 6,525,168 and 6,723,827)

v.

JONATHAN D. **ZOOK**, SUSAN E. DEMOSS,  
DAVID W. JORDAN AND CHANDRA B. RAO  
Senior Party ("PRC")  
(U.S. Application 10/644,389).

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Patent Interference No. 105.555 (MPT)  
(Technology Center 1700)

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**JUNIOR PARTY ZOOK'S PRELIMINARY MOTION LIST**

1 Pursuant to the requirements of the Declaration of this interference (Paper No. 1, page 2).  
2 and in accordance with 37 C.F.R. § 41.120 and ¶ 204 of the Standing Order (Paper No. 2, page  
3 58), the Junior Party Zook *et al* ("PBT") hereby requests the Board's authorization to file the  
4 following motions seeking the relief set forth below:

5 1. PBT Motion 1 - Pursuant to 37 C.F.R. § 41.121(iii) and 41.208(a)(1), a motion for  
6 judgment of no interference-in-fact as to all claims of Junior Party PBT U.S. Patent No.  
7 6,723,827, as all the claims of the '827 patent are directed to a separate patentable invention.

8 The interference Declaration identified all claim of the '827 patent as corresponding to  
9 the count. The count is directed to a polythioether. The '827 patent, however, was based upon a  
10 divisional application of the '168 patent is directed entirely at a process for forming a  
11 polythioether. As a divisional application, the process claims of the '827 patent cannot, by  
12 definition, be directed to the same patentable invention as the product claim of the count. Senior  
13 Party PRC U.S. Application 10/302,177 is also not enabling for the claims of the '827 patent.

14 2. PBT Motion 2 - Pursuant to 37 C.F.R. § 41.121(iii) and 41.208(a)(1), a motion for  
15 judgment that claims 22 – 27 and 35 – 43 of Senior Party PRC U.S. Application 10/302,177, that  
16 correspond to the count, are unpatentable for new matter in view of 35 U.S.C. § 112, ¶ 1.

17 35 U.S.C. § 132 provides, "... No amendment shall introduce new matter into the  
18 disclosure of the invention." The Senior Party's specification, and the claims as originally filed,  
19 recite that "m" is a rational number. In the preliminary amendment dated March 16, 2004 claim  
20 22 was amended to state that "m" "an integer between 0 and 10". It is axiomatic that "0" is an  
21 integer, but not a rational number. Therefore, the amendment of March 16, 2006 introduced new  
22 matter.

3. PBT Motion 3 - Pursuant to 37 C.F.R. § 41.121(iii) and 41.208(a)(1), a motion for judgment that claims 22 – 27 and 35 – 43 of Senior Party PRC U.S. Application 10/302,177, that correspond to the count, are unpatentable for lack of written description support as required by 35 U.S.C. 112, ¶ 1.

The Senior Party's specification, and the claims as originally filed, recite that "m" is a rational number. In independent claim 22, the term "m" is defined as "an integer between 0 and 10". Accordingly, there is no support in the '177 application for the term "m" to be defined as an "integer".

4. PBT Motion 5 - Pursuant to 37 C.F.R. § 41.121(i) and 41.208(a)(2), a motion designating claims 7, 9 and 10 -13 of Junior Party PBT U.S. Patent No. 6,525,168 as not corresponding to the count.

The count, if treated as prior art, would not have anticipated or rendered obvious the subject matter of claims 7, 9 and 10 – 13 of the '168 patent.

5. PBT Motion 6 - Pursuant to 37 C.F.R. § 41.208(a)(1), and contingent on the grant of motions 1 or 2 and the initiation of settlement discussions, a motion to add inventors Willard, Jones and Cosman of Junior Party PBT U.S. Patent Nos. 6,525,168 and 6,723,827 to of Senior Party PRC U.S. Application 10/302,177.

Respectfully submitted,

Dated: August 22, 2007

s/Mark D. Schneider  
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Counsel for Junior Party Zook

Filed 29 August 2007

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UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

JONATHAN D. **ZOOK**, DAVID W. JORDAN, DEAN M. WILLARD,  
GEORGE JONES, and MICHAEL COSMAN,  
Junior Party ("PBT")  
(U.S. Patents 6,525,168 and 6,723,827),

v.

JONATHAN D. **ZOOK**, DAVID W. JORDAN,  
SUSAN E. DEMOSS and CHANDRA B. RAO  
Senior Party ("PRC")  
(U.S. Application 10/644,389).

Patent Interference No. 105,555 (MPT)  
(Technology Center 1700)

Order Motion Times - Bd.R. 104(c)

A. Conference call

A telephone conference call was held on August 28, 2007 at approximately  
2:00 p.m., involving:

1. Mark D. Schneider, counsel for Zook PBT,
2. Jerry D. Voight, counsel for Zook PRC, and
3. Michael Tierney, Administrative Patent Judge.

1 B. Relevant discussion during conference call

2  
3 The principal purpose of the conference call was to set times for taking  
4 action on motions in the interference. Based upon the conference call the  
5 following came to light:

6 1) Junior party PBT will not be filing a brief on priority of invention.  
7 Junior party stated that it does not contest priority;

8 2) Senior Party PRC's involved '389 application lacks continuity with its  
9 parent application as PRC failed to specifically file an extension of time in the  
10 parent application prior to its abandonment;

11 3) Senior Party PRC has a reissue application that is a constructive  
12 reduction to practice of Count 1 that is prior to PBT, and contains claims that  
13 interfere with PBT. (See Order, Paper 3).

14 In light of the above facts, PBT was authorized to file a motion seeking to  
15 undesignate certain claims of its '168 patent and a motion alleging no  
16 interference-in-fact between its involved '827 patent and PRC's '389 application.  
17 PRC was authorized to file motions seeking to correct continuity and add a  
18 divisional reissue application to the interference. No other motions were  
19 authorized. A discussion of these as well as the other motions identified on the  
20 parties' lists is provided below.

21  
22 PBT's Motions List

23 1. Motion for No Interference-in-Fact

24 PBT alleges that PBT's '827 patent does not contain a claim that interferes  
25 with the claims of PRC's involved '389 application. In particular, PBT alleges that  
26 its '827 patent claims a process that is patentably distinct from the '389 patent,

1 which claims a product. The Board authorized PBT's requested no interference-in-  
2 fact motion.

3  
4 2. Motion to Undesignate PBT Claims

5 PBT requests authorization to file a motion to undesignate claims 7 and 9-13  
6 of its involved '168 patent. PBT request is authorized.

7  
8 3. Motion Alleging Lack of Written Description

9 PBT requests authorization to file a motion alleging that some, but not all,  
10 PRC claims lack written description and/or are based upon new matter.  
11 Specifically, PBT states that PRC's application describes "m" as a rational number  
12 but that PRC's claims were amended to recite that m is an integer between 0 and  
13 10. PBT states that 0 is not a rational number and concludes that the PRC claims  
14 that identify "m" as including 0 lack written description.

15 PBT does not contest priority of invention. PBT acknowledged that there is  
16 an interference-in-fact between PRC and PBT even if the "m=0" PRC claims were  
17 held unpatentable. Accordingly, the Board denies PBT's request. PRC shall, upon  
18 resumption of ex parte prosecution, inform the Examiner of PBT's rational number  
19 allegation and/or amend the claims to correct the alleged problem.

20  
21 4) Motion to Correct Inventorship

22 PBT requested authorization to correct inventorship should it be necessary.  
23 PBT's request to file motions correcting inventorship is dismissed without  
24 prejudice.

1           PRC's Proposed Motions

2           1.     Motions to Substitute a Count and Attack Benefit

3           PRC requests authorization to file a motion to substitute a count. PRC also  
4 requested authorization to file a motion attacking PBT's accorded priority benefit.  
5 PRC conceded that these motions are moot as PBT does not contest priority of  
6 invention. PRC's requests to file motions substituting a count and attacking  
7 priority benefit are dismissed without prejudice.

8  
9           2.     Motions for Unpatentability: Prior Art & Derivation

10          PRC requests authorization to file motions alleging that all of PBT's  
11 involved claims are unpatentable based upon prior art and derivation. PRC  
12 conceded that these motions are moot as PBT does not contest priority of  
13 invention.

14          PBT's only authorized motions seek to restrict the number of PBT claims  
15 involved in the interference. PBT does not contest priority of invention as to the  
16 claims that are found to correspond to Count 1. Accordingly, the Board dismissed  
17 PRC's request to file motions based upon prior art and derivation as judgment will  
18 be awarded against PBT's claims that correspond to Count 1, i.e., PBT's claims  
19 will be held unpatentable under 35 U.S.C. §102(g).

20  
21          3.     Motions to Correct Continuity

22               a.     Miscellaneous Motion for Accept Unintentionally Delay Claim  
23                       for 35 U.S.C. § 120 benefit

24  
25          PRC requests authorization to file a motion to accept an unintentionally  
26 delayed claim for 35 U.S.C. § 120 benefit of an earlier filed application. In

1 particular, PRC seeks to amend its involved '389 application to properly claim  
2 benefit of an additional application, U.S. Application 09/318,500. The Board  
3 authorizes PRC's request. The Board informed PRC however, that for purpose of  
4 the interference the Board may determine that PRC's requested relief is moot in  
5 light of PBT's failure to contest priority of invention.

6 NO OPPOSITION to PRC's miscellaneous motion to accept unintentionally  
7 delayed claim for benefit is authorized at this time.

8  
9 b. Miscellaneous Motion to Correct Copendency

10 PRC's involved '389 application claims 35 U.S.C. § 120 benefit of a chain  
11 of applications. PRC's '389 application is a continuation of U.S. Application  
12 10/302,177. PRC's '389 application was filed on August 19, 2003. PRC's parent  
13 '177 was abandoned on May 20, 2003 for failure to respond to a non-final office  
14 action. PRC apparently failed to file a request for a three-month extension of time  
15 in the '177 application prior to its filing of the child '389 application.

16 PRC's motion shall include a petition seeking the revival of its '177  
17 application for the purpose of filing a three-month extension of time such that the  
18 '177 application would have been copending with PRC's '389 application.

19 PRC's motion shall identify all necessary evidence sufficient to demonstrate  
20 that PRC intended to properly file a three-month extension of time in the '177  
21 application prior to its abandonment. Further, the Board requests that, to the extent  
22 possible, PRC provide a declaration from the attorney who filed the child '389  
23 application, with the declaration explaining how the error in copendency arose.  
24 PRC shall file any and all evidence relied upon in its motion and petition  
25 concurrent with their filing.

1 PRC requested and PBT did not oppose the expedited filing of this motion.  
2 PRC's motion is due **October 9, 2007**. Prior to filing its motion, PRC shall  
3 provide a copy of the motion to PBT in sufficient time such that PBT may  
4 determine whether or not PBT intends to oppose the motion. The first page of  
5 PRC's motion shall state whether or not PBT intends to oppose the grant of the  
6 motion. Concurrent with the filing of the motion, PRC shall file any and all  
7 evidence relied upon in the motion.

8  
9 4. Motion to Add Reissue Application

10 As discussed in the Order, Paper 3, PRC has a reissue application,  
11 10/788,965, that contains claims that interfere with PBT's involved claims. The  
12 reissue application is currently suspended awaiting the filing of a divisional reissue  
13 application where the divisional application would be directed to the interfering  
14 subject matter. PRC requests authorization to file a motion seeking to add a yet to  
15 be filed divisional reissue application to the interference.

16 PRC's reissue application is a "great-grandparent" application of the  
17 involved '389 application. A divisional of this reissue application would be  
18 accorded the same constructive reduction to practice benefit as the '389 application  
19 was accorded. The divisional reissue application would allegedly avoid the  
20 copendency problem presented by the '389 application.

21 The Board authorizes the filing of a motion to add a PRC divisional reissue  
22 application to the interference. The Board authorizes PRC to file its motion on an  
23 expedited basis on **October 9, 2007**. Should PRC file its motion on an expedited  
24 basis, PRC shall provide a copy to in sufficient time for PBT to determine whether  
25 PBT intends to oppose the motion. The first page of PRC's motion shall state

1 whether PBT intends to oppose the motion. Concurrent with the filing of the  
2 motion, PRC shall file any and all evidence relied upon in the motion.

3 PRC agreed to provide the Board with a status update on the '965 reissue  
4 application, and any divisionals thereof, on **September 29, 2007**.

5  
6 **C. Time periods associated with motions**

7 In accordance with discussion during the telephone conference call, the  
8 TIME PERIODS described below are set out in an Appendix to this ORDER.  
9 Action specified for each TIME PERIOD must be completed by the date specified  
10 for the TIME PERIOD

11 The parties are authorized to stipulate different times (earlier or later, but not  
12 later than TIME PERIOD 7) for TIME PERIODS 1 through 6.<sup>1</sup> A notice of the  
13 stipulation must be promptly filed. The notice must be in the form of a photocopy  
14 of the Appendix attached to this ORDER with old dates crossed out and new dates  
15 inserted by hand. The parties may not stipulate an extension of TIME PERIODS  
16 7-9.

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<sup>1</sup> In stipulating different times, the parties should consider the effect of the stipulation on times (1) to object to evidence (5 business days, Bd.R. 155(b)(1)), (2) to supplement evidence (10 business days, Bd.R. 155(b)(2)), (3) to begin cross examination (no earlier than 21 days after service, SO & 157.3.1) and (4) to conclude cross examination (at least 10 days before the opposition or reply is due, SO & 157.3.2).

1  
2  
3  
4

**1. TIME PERIOD 1**

- a. File and serve all authorized motions and
- b. Serve but do not file evidence in support of these motions.

If no party files a motion, the SENIOR PARTY must arrange a conference call with the parties and the Board so that appropriate adjustments to the schedule may be made.

**2. TIME PERIOD 2**

- a. File and serve responsive motions (Bd.R. 121(a)(2)) in response to an opponent's motion filed during TIME PERIOD 1 and
- b. Serve but do not file evidence in support of these responsive motions.

**3. TIME PERIOD 3**

- a. File and serve oppositions to all motions, including responsive motions and
- b. Serve but do not file evidence in support of these oppositions.

**4. TIME PERIOD 4**

- a. File and serve replies to all oppositions and
- b. Serve but do not file evidence in support of these replies.

**5. TIME PERIOD 5**

- a. File and serve any request for oral argument on motions,
- b. File and serve motions to exclude evidence (Bd.R. 155(c); SO & 155.2), and

- c. File and serve observations on cross examination (SO & 157.7) of reply testimony.

**6. TIME PERIOD 6**

- a. File and serve oppositions to an opponent's motion to exclude evidence and
- b. File and serve any response to observations.

**7. TIME PERIOD 7**

File and serve replies to oppositions to motions to exclude evidence.

**D. Deposition transcripts**

Transcripts of cross examinations and depositions taken under 35 U.S.C. 24 must be served, but not filed until the exhibits are filed.

**E. Serving exhibits relied upon in motions**

An exhibit, including an affidavit, cited in connection with a motion, opposition, reply, or affidavit, must be served. but not filed,<sup>2</sup> with the motion, opposition, reply or affidavit in which the exhibit is first mentioned.

**F. TIME PERIOD 8: Filing the record for decision on motions**

- 1. File an original set of your exhibits and one working copy of your exhibits;

---

<sup>2</sup> Except when the Board sets an expedited schedule for a particular motion, in which case, all exhibits mentioned in that motion or the corresponding opposition or reply must be filed with the motion, opposition, reply, or affidavit in which the exhibit is first mentioned.

2. For each of your motions, file one folder (or three folders if an oral argument is set each) containing:
  - a. The motion,
  - b. Any corresponding opposition,
  - c. Any corresponding reply,
  - d. Any corresponding observations, and
  - e. Any corresponding response to the observations.
3. File CD-ROM a party elects to file.

#### **G. TIME PERIOD 9: Default oral argument date**

If a request for oral argument (Bd.R. 124(a); TIME PERIOD 5) is granted, the default date for such argument is TIME PERIOD 9. No oral argument will occur if either no argument is requested or granted.

#### **H. Priority statements**

1. At TIME PERIOD 1:
  - a. File but do not serve a priority statement (Bd.R. 120; Bd.R. 204(a)).
  - b. File and serve a notice advising each opponent of the filing of the priority statement.
2. A junior party who does not file a priority statement shall not have access to the priority statement of any other party.

3. **Within one (1) week** after TIME PERIOD 1, serve a copy of the priority statement upon each opponent (except for a junior party barred under & H.2 above).

cc (via electronic filing):

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Revised 3 January 2006

**Appendix--ORDER - RULE 123(a)**  
**(Times for substantive motions; priority deferred)**

Interference 105,555 (MPT)

TIME PERIOD 1 ..... **November 9, 2007**  
    File motions  
    File (but serve one week later) priority statements

TIME PERIOD 2 ..... **November 30, 2007**  
    File responsive motions to motions  
    filed in TIME PERIOD 1

TIME PERIOD 3 ..... **January 11, 2008**  
    File oppositions to all motions

TIME PERIOD 4 ..... **February 22, 2008**  
    File all replies

TIME PERIOD 5 ..... **April 4, 2008**  
    File request for oral argument  
    File motions to exclude  
    File observations

TIME PERIOD 6 ..... **April 28, 2008**  
    File oppositions to motions to exclude  
    File response to observations

TIME PERIOD 7 ..... **May 8, 2008**  
    File replies to oppositions to motions to exclude

TIME PERIOD 8 ..... **May 15, 2008**  
    File exhibits  
    File sets of motions  
    File any CD-ROMs

TIME PERIOD 9 ..... **June 13, 2008**  
    Default oral argument date (if ordered)

Filed on behalf of: Senior Party Jonathan D. Zook

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Patent Interference No. 105,555 (MPT)  
(Technology Center 1700)

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**SENIOR PARTY ZOOK (PRC) MOTION 2**  
(Miscellaneous Motion Requesting Acceptance of an  
Unintentionally Delayed Request for Priority Benefit)

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1 **I. PRECISE RELIEF REQUESTED**

2 Senior Party Zook (PRC) moves under 37 C.F.R. § 41.121(a)(3) to perfect the previously  
3 claimed 35 U.S.C. § 120 benefit of priority of U.S. Patent Application Nos. 08/802,130 and  
4 09/318,500, now U.S. Patent Nos. 5,912,319, and 6,232,401, respectively. Specifically, PRC  
5 seeks to amend the specification of involved Application No. 10/644,389 to properly recite the  
6 alternate priority chain with the correct relationships between applications to perfect a claim for  
7 35 U.S.C. § 120 benefit of these earlier applications. If necessary, PRC also seeks to amend the  
8 '389 application's parent, now abandoned Application No. 10/302,177.

9 **II. EVIDENCE IN SUPPORT OF THE MOTION**

10 In accordance with ¶121.5.1 of the Standing Order, a list of exhibits cited in support of  
11 this motion is attached as Appendix 1.

12 **III. STATEMENT OF MATERIAL FACTS**

13 In accordance with ¶121.5.2 of the Standing Order, and the Order filed August 29, 2007  
14 [Paper No. 22], PRC's statement of material facts supporting this motion is attached as Appendix  
15 2. PRC's material facts will be referred to as "MF" in this motion.

16 **IV. REASONS FOR GRANTING RELIEF REQUESTED**

17 **A. Introduction**

18 The present interference was declared on the basis of a single count, Count 1,  
19 corresponding to claim 6 of U.S. Patent 6,525,168. Paper No. 1, p. 5. In the Declaration of  
20 interference, PRC's involved Application No. 10/644,389 ("the '389 application") was accorded  
21 the benefit of four (4) prior-filed applications for Count 1, whereby PRC was accorded an  
22 effective filing date of February 19, 1997. Paper 1, p. 6. Upon review of the file histories of the  
23  
24

1 involved '389 application and the four prior-filed applications to which PRC is accorded benefit,<sup>1</sup>  
2 PRC discovered that a previously claimed benefit of priority through an alternative chain of  
3 applications that includes an application which is not part of the basis for accorded benefit in the  
4 interference, includes errors in the relationships between applications. MFs 4-6.

5 The somewhat complex priority chains relevant to the involved '389 patent are easily  
6 understood by reference to Exhibit 1003. Exhibit 1003 shows all relevant parent applications,  
7 numbered in order of filing dates, and their relationships. In Exhibit 1003, the proper priority  
8 benefit chains are shown connected by solid lines, while the improper alternative priority chain as  
9 recited in the '389 application is shown illustrated by a line composed of dashes.

10 After declaration of the present interference, PRC became aware that the alternate priority  
11 chain recited in parent U.S. Application No. 10/302,177 ("the '177 application") is ambiguous  
12 while the alternate priority chain recited in the involved '389 application improperly indicates that  
13 the application is a continuation-in-part (CIP) of U.S. Patent Application No. 09/318,500 ("the  
14 '500 application"), an application not included in the application to which movant was accorded  
15 priority benefit. MFs 5-6. However, there was not copendency between the '500 application,  
16 which issued as U.S. Patent 6,232,401 on May 15, 2001, and either the '177 application, filed on  
17 November 21, 2002, or the '389 application, filed on August 19, 2003; neither of these  
18 applications can be a CIP of the '500 application. MF 7. Rather, in accordance with the alternate  
19 priority claim, the '177 application is a continuation of another parent application, U.S. Patent

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21 <sup>1</sup> Specifically, PRC was accorded benefit of the filing dates of prior U.S. Patent Application Nos.  
22 10/302,177, 09/756,573, 08/928,972 ("the '972 application"), and 08/802,130 ("the '130  
23 application").  
24

1 Application No. 09/756,573 ("the '573 application"), which is a CIP of the '500 application,  
2 while the '389 application is a continuation of the '177 application. MF 8.

3 PRC seeks to perfect the '389 application's alternate priority claim by amending the first  
4 paragraph of the '389 application to properly recite the relationship between the '389 application  
5 and the '500 application. This previously authorized miscellaneous motion requests the Board to  
6 accept an unintentionally delayed claim for 35 U.S.C. § 120 benefit of an earlier filed application.

7 **B. PRC is Entitled to Claim the Benefit of the '500 Application (No. 3)**

8 Requirements sufficient for claiming the benefit of an earlier filing date are set forth in  
9 part in 37 C.F.R. § 1.78(a). MF 9. This rule imposes six requirements which must be met in  
10 order for an application to receive the benefit of an earlier-filed application's filing date.  
11 Specifically, 37 C.F.R. § 1.78(a) requires: (i) copendency; (ii) common inventorship; (iii)  
12 compliance with 35 U.S.C. § 112, first paragraph; and (iv) compliance with Rule 1.53(b) or (d);  
13 and 37 C.F.R. § 1.78(b) requires: (v) reference in the application to each prior-filed application,  
14 identifying the prior application by application number and indicating the relationship of the  
15 applications, (vi) which reference must be submitted within the later of four months from the  
16 actual filing date of the later-filed application or sixteen months from the filing date of the prior-  
17 filed application. Only the fifth and sixth requirements are in issue with regard to the alternative  
18 priority claim. MF 10.

19 The first requirement of Rule 1.78, copendency, is satisfied. MFs 9 & 11. The '389  
20 application (No. 6) was filed on August 19, 2003, as a continuation of the '177 application (No.  
21 5). The '177 application (No. 5) was filed on November 21, 2002, as a continuation of the then-  
22 pending '573 application (No. 4), which did not issue until January 21, 2003. The '573  
23 application (No. 4) was filed on January 8, 2001, as, *inter alia*, a CIP of the then-pending '500  
24

1 application (No. 3), which did not issue until May 15, 2001. The '500 application (No. 3) was  
2 filed on May 25, 1999, and was a divisional of the then-pending '130 application (No. 1), which  
3 did not issue until June 15, 1999. Thus, effective copendency for purposes of claiming priority  
4 existed.

5 The second requirement of Rule 1.78 is that the later filed application must have at least  
6 one inventor in common with the prior-filed application. MF 9. Each application of the alternate  
7 priority chain, the '130, '500, '573, '177, and '389 applications names, *inter alia*, Jonathan D.  
8 (Doherty) Zook, Susan E. DeMoss (formerly known as Suzanna Gibson DeMoss and Suzanna  
9 Gibson), Chandra B. Rao, and David W. (Weldon) Jordan. MF 12. Accordingly, the common  
10 inventor requirement of § 1.78(a)(1) is satisfied.

11 The third requirement of Rule 1.78 is that the prior filed application must disclose the  
12 invention claimed in at least one claim of the later filed application in accordance with the first  
13 paragraph of section 112. MF 9. Pending claims in the '389 application (No. 6) have been  
14 accorded the benefit of the '130 application. Paper 1, pp. 5-6. This benefit flowing from the '130  
15 application through the '972, '573, and '177 applications to the '389 application is unchallenged.  
16 MF 13. The '500 application is a divisional of the '130 application having an identical  
17 specification. MF 14. As such, the '500 application discloses each invention disclosed in the  
18 '130 application and must also, therefore, disclose the invention claimed in at least one claim of  
19 the later filed application, thereby satisfying the disclosure requirement of § 1.78(a)(1).

20 The fourth requirement of Rule 1.78 is that the prior application must be entitled to a filing  
21 date as set forth in § 1.53(b) or § 1.53(d) and include the basic filing fee as set forth in § 1.16.  
22 MF 9. The '500 application (No. 3) was filed as a divisional of the '130 application (No. 1) under  
23 37 C.F.R. § 1.53(b). MF 14. The statutory filing fee and surcharge, with authorization to charge  
24

1 any additional amounts due, was paid in the Response to Notice to File Missing Parts, filed July  
2 16, 1999, within the two month deadline. MF 15. Accordingly, the requirement that the prior  
3 application be entitled to a filing date is satisfied.

4 The fifth requirement of Rule 1.78 is that the later-filed application must reference each  
5 prior-filed application, identifying each by application number and indicating the relationship of  
6 the applications as set forth in § 1.78(b). MF 9. This requirement has not been fully met. The  
7 first paragraph of the '389 application (No. 6) was amended to recite the '500 application (No. 3)  
8 by its application number and indicates that the '389 application is a CIP of the '500 application.  
9 MF 16. While accurately setting forth the application number and correctly characterizing the  
10 nature of the applications' disclosures, the recited relationship between the '389 and '500  
11 applications is not correct. MF 17. Similarly, the relationship between the '177 and '500  
12 applications recited in the '177 application is ambiguous and not necessarily correct. MF 18.

13 The sixth requirement of Rule 1.78 is that the required reference to each prior-filed  
14 application must be made within the later of four months from the actual filing date or sixteen  
15 months from the filing date of the prior-filed application. MF 9. Reference to the prior-filed '500  
16 application (No. 3) in the '389 application (No. 6) was made within four months of the actual  
17 filing date by, *inter alia*, amendment of the specification at the time of filing to recite "[t]his  
18 application [the '389 application] is ... also a continuation-in-part of U.S. Patent Application No.  
19 09/318,500." MF 19. Exhibit 1006, paragraph 9, page 2. Similarly, the specification of the '177  
20 application was amended at the time of filing to recite "[t]his application is ... also a  
21 continuation-in-part of U.S. Patent Application No. 09/318,500" MF 20. Exhibit 1007, paragraph  
22 9, page 2. However, the amendments to the specifications did not clearly set forth the relationship  
23 of the relevant application.

1 Thus, it is only through error in the recited relationship between the '389 application and  
2 the '500 application and between the '177 application and the '500 application that the '389 and  
3 '177 applications would not be entitled to claim priority to the '130 application through a priority  
4 chain that involves the '500 application.

5 **C. PRC is Entitled to Perfect the Alternate Priority Benefit Chain**

6 **1. Amendment of Priority Claim is Proper where Delay is Unintentional**

7 Requirements sufficient for claiming the benefit of an earlier filing date after the time  
8 period specified by § 1.78(a)(2), namely, later than four months from the filing date of the later-  
9 filed application and sixteen months from the filing date of the prior-filed application, are set  
10 forth in part in 37 C.F.R. § 1.78(a)(3). MF 21. This rule imposes three requirements to allow an  
11 unintentionally delayed claim for the benefit of priority under 35 U.S.C. § 120. Specifically, it  
12 requires that a petition be accompanied by: (i) the required reference to the prior-filed application;  
13 (ii) the surcharge set forth in § 1.17(t); and (iii) a statement that the entire delay from the date the  
14 claim was due has been unintentional. Petitions for the '389 and '177 applications are attached  
15 (Appendices 3 and 5, respectively).

16 The first requirement for a grantable petition under Rule 1.78(c) is the required reference  
17 to the prior-filed application. MF 21. The required reference is provided for the '389 application  
18 (No. 6) by amendment (Appendix 4). If the Board determines that the '177 application (No. 5)  
19 should similarly be amended, PRC petitions the Board under 37 C.F.R. § 1.78 and/or § 1.182 to  
20 enter the corresponding amendment for that case (Appendix 6). MF 22. These amendments each  
21 correct the improperly recited relationship between the applications as discussed above such that  
22 the alternate priority chain flows correctly from the '130 application through the '500, '573, and  
23 '177 application to the '389 application as set forth by the solid line in Exhibit 1003.

1 The second requirement under Rule 1.78(c), the required surcharge as set forth in  
2 § 1.17(t), is met by authorization to charge a deposit account provided herewith (\$1,410) for both  
3 the '389 application (MF 23) and for, if deemed necessary, the '177 application (MF 24).

4 The third requirement under Rule 1.78(c), a statement that the entire delay from the date  
5 the claim was due has been unintentional, is also provided herewith for both the '378 application  
6 (No. 6) and for the '177 application (No. 5).

7 Thus, the petition for acceptance of Applicants' unintentionally delayed claim for the  
8 benefit of priority under 35 U.S.C. § 120 is a grantable petition for both the '389 application and,  
9 if deemed necessary, the '177 application, which would perfect the alternative priority claim.

10 **2. The Evidence Unambiguously Establishes that the Entire Period of**  
11 **Delay in Remedying the Error in the Priority Claim was Unintentional**

12 The only real issues presented by the present motion are whether the failure to properly  
13 recite the relationship between the '389 application, and if it is necessary for the grant of relief,  
14 the '177 application, and the other applications of the alternate priority chain that flows through  
15 the '500 application was unintentional and remained so until presentation of the present motion  
16 and petition. The facts indicate that the original error and the entire delay in correcting the error  
17 in the recited relationship between applications was unintentional.

18 The first paragraph of the '573 application (No. 4), as filed on January 8, 2001, states:

19 This application is a continuation-in-part of U.S. Patent Application  
20 No. 08/928,972 filed September 12, 1997, which is a continuation-  
21 in-part of now U.S. Patent No. 5,912,319. Also, this application is a  
22 continuation-in-part of U.S. Patent Application No. 09/318,500  
23 filed May 25, 1999, which is a division of now U.S. Patent No.  
24 5,912, 319. This application also claims the benefit of U.S.  
provisional application no. 60/182,396 filed February 14, 2000 and  
U.S. provisional application no. 60/215,548 filed June 30, 2000.

1 MF 25. Upon receiving a corrected filing receipt with errors in, *inter alia*, the domestic priority  
2 data as claimed by Applicants, a corrected filing receipt was requested. MF 26. The Request for  
3 Corrected Filing Receipt again recited, *inter alia*, the correct relationship between the '500 and  
4 other applications, namely, "[t]his application [the 09/756,573 application] is also a CIP of  
5 09/318,500 05/25/1999 PAT 6,232,401 which is a DIV of 08/802,130 02/19/97 PAT 5,912,319."  
6 MF 26. This relationship is correctly recited in the first paragraph of U.S. Patent No. 6,509,418,  
7 which issued from the '573 application on January 21, 2003. Exhibit 1014.

8 On November 21, 2002, the '177 application (No. 5) was filed as a continuation of the  
9 '573 application (No. 4). MF 27. The application transmittal directed the amendment of the  
10 specification to recite the claimed benefit of priority; the directed amendment recited:

11 This is a continuation of U.S. Patent Application No. 09/756,573,  
12 filed January 8, 2001, which is a continuation-in-part of U.S. Patent  
13 Application No. 08/928,972, filed September 12, 1997, which is a  
14 continuation-in-part of now U.S. Patent No. 5,912,319; and also a  
15 continuation-in-part of U.S. Patent Application No. 09/318,500,  
16 filed May 25, 1999, which is a division of now U.S. Patent No.  
17 5,912,319; and claims the benefit of U.S. Provisional Application  
18 No. 60/182,396, filed February 14, 2000, and U.S. Provisional  
19 Application No. 60/215,548, filed June 30, 2000, all of which are  
20 incorporated herein by reference.

21 Exhibit 1007, paragraph 9, page 2. As written, the directed amendment was intended to convey  
22 that the '177 application (No. 5) was a continuation of the '573 application (No. 4) and that the  
23 '573 application was a continuation-in-part of the '972 application (No. 2) and a continuation-in-  
24 part of the '500 application (No. 3). MF 28. As stated by both Messers Makrogiannis and Uhl in  
their declarations (Exhibit 1027, ¶¶ 13, 15-17, and 19, and Exhibit 1023, ¶¶ 33-34, 37, 39, and 41),  
they did not realize that the recited claim to priority in the '177 application (No. 5) was  
ambiguous at the time it was initially made or at any time afterward until being informed of such  
by interference counsel. MF 29. As stated by Mr. Lambert in his declaration (Exhibit 1022,

¶¶28-29), when he became involved with the prosecution of the then pending '177 application (No. 5), he noticed a discrepancy between the directed amendment to the priority claim, but did not notice that the priority chain recited was ambiguous. Exhibit 1022, ¶¶32, 34, and 36. MF 30. As they were at all times unaware of the ambiguity in the priority claim, the failure to correct the recited relationship between the applications in the priority chain was and remained unintentional at all times.

On August 19, 2003, the '389 application (No. 6) was filed as a continuation of the '177 application (No. 5). As stated by Mr. Lambert in his declaration (Exhibit 1022, ¶30), he filed the '389 application (No. 6) with some changes in punctuation to the priority claim, but did not realize that the alternative priority chain could be construed to incorrectly suggest the '389 application (No. 6) claimed priority directly to the '500 application (No. 3). Exhibit 1022, ¶31. While he noticed some discrepancies between the directed amendment and what was recited in the original Filing Receipt, and he requested a Correct Filing Receipt, he did not appreciate any deficiency in the alternative priority chain. Exhibit 1022, ¶33. Further, at no time was he aware of any deficiency in the priority claims involving the '500 application (No. 3) until being informed of such deficiencies by interference counsel. Exhibit 1022, ¶34. MF 30.

As stated by Mr. Uhl in his declaration, he also did not realize that the alternative priority chain could be construed to incorrectly suggest the '389 application (No. 6) claimed priority directly to the '500 application (No. 3) (Exhibit 1022, ¶36), did not appreciate any deficiency in the alternative priority chain when Mr. Lambert obtained a corrected filing receipt (Exhibit 1022, ¶38), and was at no time aware of any deficiency in the priority claims involving the '500 application (No. 3) until being informed of such deficiencies by interference counsel (Exhibit 1022, ¶39). MF 29.

1 Moreover, not being the basis for a benefit claim sought in the present interference,  
2 nothing occurred during the prosecution of related applications that alerted them to any deficiency  
3 in the alternative priority claim. The sole priority issue that arose during prosecution of the '573,  
4 '177, and '389 applications was whether, during prosecution of the '389 application, there was  
5 support for the claims in the '389 application in the '130 application. MF 31.

6 If no one was aware of any error in the recited relationship between applications until very  
7 recently, where each and every application was identified by application number and the nature of  
8 the applications' disclosure relative to one another was correctly set forth, how could the  
9 ambiguity or error in the recited relationship between applications be anything but unintentional?

10 **V. CONCLUSION**

11 For the reasons set forth above, PRC respectfully requests that the Board grant this motion  
12 and associated petition (Appendix 3) to accept an unintentionally delayed claim for 35 U.S.C.  
13 § 120 benefit for the '389 application, enter the amendment setting forth the proper alternate  
14 priority claim (Appendix 4), and, if necessary to amend the '177 application (No. 5), the related  
15 petition (Appendix 5) under Rules 1.78 and/or 1.182 to enter the corresponding amendment for  
16 that case (Appendix 6), to perfect the alternative priority benefit claim involving Application No.  
17 09/318,500 (No. 3).

18  
19 Date: November 30, 2007

Respectfully submitted,

20 By: /Jerry D. Voight/  
21 Jerry D. Voight  
Registration No. 23,020

22 Counsel of Record for Senior Party Zook  
23  
24

## **APPENDIX 1**

### **List of Exhibits**

- 1003:** Chart captioned "Senior Party Zook (PRC) Involved Application Family Tree."
- 1006:** Application Transmittal for Application No. 10/644,389, filed August 19, 2003.
- 1007:** Application Transmittal for Application No. 10/302,177, filed November 21, 2002.
- 1008:** Declaration and Power of Attorney filed for Application No. 10/302,177 on November 21, 2002.
- 1009:** Declaration and Power of Attorney filed for Application No. 09/756,573 on June 12, 2001 in Response to Notice to File Missing Parts of Application.
- 1010:** Declaration and Power of Attorney filed for Application No. 10/644,389 on August 19, 2003.
- 1014:** Copy of U.S. 6,509,418 B1, issued from Application No. 09/756,573.
- 1022:** Declaration of William R. Lambert, dated October 1, 2007.
- 1023:** Declaration of William J. Uhl, dated October 2, 2007.
- 1026:** Request for Corrected Filing Receipt for Application No. 09/756,573, filed June 12, 2001.
- 1027:** Declaration of Philippe N. Makrogiannis, dated November 8, 2007.
- 1028:** Application Transmittal for Application No. 09/318,500, filed May 25, 1999.
- 1029:** Copy of U.S. 6,232,401, issued from Application No. 09/318,500.
- 1031:** Copy of Decision on Petition dated October 19, 2007, reviving Application No. 10/302,177, establishing continuity with continuing Application No. 10/644,389.
- 1032:** Application Transmittal for Application No. 09/756,573, filed January 8, 2001.
- 1033:** Copy of U.S. 5,912,319, issued from Application No. 08/802,130.
- 1034:** Copy of Response to Notice to File Missing Parts filed July 16, 1999, in response to Notice dated June 16, 1999, with a due date of August 16, 1999, for response, in connection with the '500 application.
- 1035:** Copy of Declaration for Patent Application filed with Application No. 09/756,573.

## APPENDIX 2

### Statement of Material Facts

1. Interference No. 105,555 (MPT), declared July 10, 2007, involves U.S. Patent Nos. 6,525,168 (“the ‘168 patent”) and 6,723,827 (the ‘827 patent”) and Application No. 10/644,389 (“the ‘389 application”). Paper No. 1.

2. PRC-DeSoto International, Inc. (“PRC”) is a real party in interest of the ‘389 application.

3. Advanced Chemistry and Technology, Inc. (“PBT”) is a real party in interest of the ‘168 and ‘827 patents.

4. In declaring the interference, the Board accorded PRC benefit for Count 1 of Application No. 10/644,389, filed August 19, 2003 (“the ‘389 application”), Application No. 10/302,177, filed November 21, 2002 (“the ‘177 application”), Application No. 09/756,573, filed January 8, 2001 (“the ‘573 application”), issued as US Patent No. 6,509,418 on January 21, 2003, Application No. 08/928,972, filed September 12, 1997 (“the ‘972 application”), issued as US Patent No. 6,172,179, and Application No. 08/802,130, filed February 19, 1997 (“the ‘130 application”), issued as US Patent No. 5,912,319. Paper No. 1, p. 6. The relationship between these applications is shown in Exhibit 1003, a chart which also shows the relationship of Application No. 09/318,500 (“the ‘500 application”).

5. As recited in the alternate priority chain, the relationship between the ‘500 application and the ‘177 application is ambiguous. Exhibit 1007.

6. As recited in the alternate priority chain, the relationship between the ‘500 application and the ‘389 application is incorrect as it improperly indicates that the ‘389 application is a continuation-in-part of the ‘500 application. Exhibit 1006.

7. Copendency requires that applications be pending, neither abandoned, nor issued as a patent, on the same date. As the ‘500 application issued as U.S. Patent 6,232,401 on May 15, 2001, and the ‘177 application was not filed until November 21, 2002, and the ‘389 application was not filed until August 19, 2003; neither one of the ‘177 and ‘389 applications was copending with the ‘500 application. Exhibit 1003.

8. The first paragraph of US Patent No. 6,509,418, issued from the '573 application, indicates that the '573 application is a CIP of the '500 application. Exhibit 1014. The '177 application is a continuation of the '573 application and the '389 is a continuation of the '177 application. Exhibits 1003 and 1006-1007.

9. 37 C.F.R. § 1.78(a) requires only that any prior application relied upon for priority benefit must name at least one inventor named in the later application and that the prior application must disclose an invention of an inventor named in each prior application that is claimed in the later application and that, in addition to the requirements set forth in 35 U.S.C. § 120, an identification of each prior application and their relationship be made within the statutory time period.

10. The fifth and sixth requirements are set forth in 37 C.F.R. § 1.78 (a)(2) and provide that an application claiming filing date benefit of a prior application:

(2) ... must contain or be amended to contain a reference to each such prior-filed application, identifying it by application number ... and indicating the relationship of the applications ... [t]his reference must be submitted during the pendency of the later-filed application. If the later-filed application is an application filed under 35 U.S.C. 111(a), this reference must also be submitted within the later of four months from the actual filing date of the later-filed application or sixteen months from the filing date of the prior-filed application.

37 C.F.R. § 1.78(a)(2).

11. The '389 application (No. 6) was filed on August 19, 2003, as a continuation of the '177 application (No. 5); copendency between the applications has been established. Exhibit 1031. The '177 application (No. 5) was filed on November 21, 2002, as a continuation of the then-pending '573 application (No. 4), which did not issue until January 21, 2003. Exhibits 1007 and 1014. The '573 application (No. 4) was filed on January 8, 2001, as, *inter alia*, a CIP of the then-pending '500 application (No. 3), which did not issue until May 15, 2001. Exhibits 1014, 1032, and 1029. The '500 application (No. 3) was filed on May 25, 1999, and was a divisional of

the then-pending '130 application (No. 1), which did not issue until June 15, 1999. Exhibits 1028 and 1033. Thus, effective copendency for purposes of claiming priority existed. Exhibit 1003.

12. The '389 application was a continuation application under 37 C.F.R. § 1.53(b) filed with a declaration from, and named, the same inventors as the '177 application. Exhibits 1006, 1008, and 1010. The '177 application was a continuation application under 37 C.F.R. § 1.53(b) as a continuation of the '573 application. Exhibit 1007. The '573 application was a continuation-in-part of the '500 application, which was a divisional of the '130 application. Exhibits 1028 and 1032.

13. The benefit of priority flowing from the '130 application through the '972, '573, and '177 applications to the '389 application was accorded by the Board and is unchallenged. Paper 1, p. 6, and Paper 22, page 2.

14. The '500 application is a divisional of the '130 application, filed under 37 C.F.R. § 1.53(b), having an identical specification. Exhibit 1028.

15. The PTO issued a Notice to File Missing Parts dated June 16, 1999, with a due date of August 16, 1999, for response, in connection with the '500 application. The Applicants responded on July 16, 1999, paying the necessary filing fee and surcharge. Exhibit 1034. The PTO granted the '500 application the filing date of May 25, 1999, as indicated on the face of U.S. Patent 6,232,401, which issued from the '500 application. Exhibit 1029.

16. The transmittal for Application No. 10/644,389 directed amendment of the specification to recite a priority chain that included a claim for the benefit of the '500 application by its application number and indicates that the '389 application is a CIP of the '500 application. Exhibit 1006, paragraph 9, page 2.

17. The '389 application is a continuation of the '177 application which is a continuation of the '573 application, which is a continuation-in-part of the '500 application and also claims the benefit of priority to the recited provisional applications, 60/182,396 and 60/215,548. Exhibits 1003, 1009, 1032, and 1035.

18. The relationship between the '177 and '500 applications recited in the '177 application is ambiguous and not necessarily correct. Exhibits 1003, 1022, ¶28, 1023, ¶33, and 1027, ¶13.

19. Reference to the prior-filed '500 application (No. 3) was made in the transmittal for the '389 application (No. 6), which reference was necessarily made within four months of the actual filing date, which transmittal directed amendment of the first paragraph to recite "[t]his application [the '389 application] is ... also a continuation-in-part of U.S. Patent Application No. 09/318,500." Exhibit 1006, paragraph 9, page 2.

20. Reference to the prior-filed '500 application (No. 3) was made in the transmittal for the '177 application (No. 5), which reference was necessarily made within four months of the actual filing date, which transmittal directed amendment of the first paragraph to recite "[t]his application is ... also a continuation-in-part of U.S. Patent Application No. 09/318,500." Exhibit 1007, paragraph 9, page 2.

21. Section 1.78 (a)(3) provides in part:

If the reference required by 35 U.S.C. 120 and paragraph (a)(2) of this section is presented after the time period provided by paragraph (a)(2)ii) of this section, the claim under 35 U.S.C. 120 ... for the benefit of a prior-filed copending non-provisional application ... may be accepted if the reference identifying the prior-filed application by application number ... was unintentionally delayed. A petition to accept an unintentionally delayed claim under 35 U.S.C. 120 ... for the benefit of a prior-filed application must be accompanied by: (i) The reference required by 35 U.S.C. 120 and paragraph (a)(2) of this section to the prior-filed application, unless previously submitted; (ii) The surcharge set forth in § 1.17(t); and (iii) A statement that the entire delay between the date the claim was due under paragraph (a)(2)(ii) of this section and the date the claim was filed was unintentional. The Director may require additional information where there is a question whether the delay was intentional.

37 C.F.R. § 1.78(a)(3).

22. If the Office determines that the '177 application requires amendment and that Rule 1.78 does not specifically provide for such amendment under the present circumstances, PRC petitions the Board under 37 C.F.R. § 1.182 in accordance with the discussion in the submitted petition under Rule 1.78 (Appendix 5) to amend the '177 application as directed (Appendix 6). This amendment of an abandoned intermediate application to contain a specific reference to an earlier-filed application is in accordance with 35 U.S.C. § 120, when it is determined to be necessary. *Sampson v. Comm'r of Patents and Trademarks*, 195 USPQ 136 (D.D.C. 1976). Upon determination by the Board that this petition is necessary, the Patent Office is authorized to charge Deposit Account No. 06-0916 the requisite fee of \$1,410.00 for this petition under 37 C.F.R. § 1.17(t) as well as any additional fees that may be necessary.

23. Authorization to charge Deposit Account No. 06-0916 the requisite fee under 37 C.F.R. § 1.17(t), \$1,410.00, is provided in Appendix 3 submitted herewith.

24. Authorization to charge Deposit Account No. 06-0916 the requisite fee under 37 C.F.R. § 1.17(t), \$1,410.00, is provided in Appendix 5 submitted herewith.

25. The first paragraph of the '573 application (No. 4), as filed on January 8, 2001, states:

This application is a continuation-in-part of U.S. Patent Application No. 08/928,972 filed September 12, 1997, which is a continuation-in-part of now U.S. Patent No. 5,912,319. Also, this application is a continuation-in-part of U.S. Patent Application No. 09/318,500 filed May 25, 1999, which is a division of now U.S. Patent No. 5,912, 319. This application also claims the benefit of U.S. provisional application no. 60/182,396 filed February 14, 2000 and U.S. provisional application no. 60/215,548 filed June 30, 2000.

26. The Request for Corrected Filing Receipt, dated June 12, 2001, recited, *inter alia*, the correct relationship between the '500 and other applications, namely, "[t]his application [the 09/756,573 application] is also a CIP of 09/318,500 05/25/1999 PAT 6,232,401 which is a DIV of 08/802,130 02/19/97 PAT 5,912,319." Exhibit 1026.

27. The '177 application (No. 5) was a continuation of the '573 application (No. 4) under 37 C.F.R. § 1.53(b); the '177 application was filed on November 21, 2002. Exhibit 1007.

28. Messers Makrogiannis and Uhl intended the amendment directed in the application transmittal to convey that the '177 application (No. 5) was a continuation of the '573 application (No. 4) and that the '573 application was a continuation-in-part of the '972 application (No. 2) and the '500 application (No. 3). Exhibits 1023, ¶¶33, 37, and 40 and 1027, ¶¶13, 15-16, and 18.

29. To the best of Mr. Makrogiannis's knowledge, neither he nor anyone affiliated with PRC were aware that the alternative chain of priority, which involves the '500 application, had any deficiency until a defect was discovered by interference counsel after the interference was declared. Exhibit 1027, ¶17. To the best of Mr. Uhl's knowledge, neither he nor anyone affiliated with PRC were aware that the alternative chain of priority, which involves the '500 application, had any deficiency until a defect was discovered by interference counsel after the interference was declared. Exhibit 1023, ¶39.

30. To the best of Mr. Lambert's knowledge, neither he nor anyone affiliated with PRC were aware that the alternative chain of priority, which involves the '500 application, had any deficiency until a defect was discovered by interference counsel after the interference was declared. Exhibit 1022, ¶34.

31. The sole priority issue that arose during the prosecution of Application Nos. 09/756,573, 10,302,177, and 10/644,389 was whether certain claims in the '573 application were supported by the specification of Application No. 08/802,130, as the USPTO considered the '389 application as a continuation-in-part of the '130 application.

**APPENDIX 3**

**PATENT**  
Customer No. 22,852  
Attorney Docket No. 08303.0042-06000

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of: )  
)  
Jonathan D. ZOOK *et al.* ) ATTN: Office of Petitions  
)  
Application No.: 10/644,389 ) Group Art Unit: 1711  
)  
Filed: August 19, 2003 ) Examiner: Duc TRUONG  
)  
For: SEALANTS AND POTTING )  
FORMULATIONS INCLUDING )  
MERCAPTO-TERMINATED )  
POLYMERS PRODUCED BY THE )  
REACTION OF A POLYTHIOL )  
AND POLYVINYL ETHER )  
MONOMER )

**Mail Stop Petition**  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

**PETITION UNDER 37 C.F.R. § 1.78(a)(3) TO ACCEPT AN  
UNINTENTIONALLY DELAYED CLAIM FOR DOMESTIC PRIORITY**

In a pending interference, *Zook v. Zook*, Patent Interference No. 105,555 (MPT), it has been noted that an alternative priority benefit chain, not the basis of priority in the interference, appears to include some errors. Applicants hereby submit this petition under 37 C.F.R. § 1.78(a)(3) to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120.

The above-referenced application includes a valid priority claim under 35 U.S.C. § 120 to a benefit chain, namely, to Application No. 10/302,177, filed November 21, 2002, now abandoned, which is a continuation of Application No. 09/756,573, filed on January 8, 2001, now U.S. Patent 6,509,418, which is a continuation-in-part of Application No. 08/928,972, filed

September 12, 1997, now U.S. Patent 6,172,179, which is a continuation-in-part of Application No. 08/802,130, filed February 19, 1997, now U.S. Patent 5,912,319.<sup>1</sup>

Application No. 10/644,389's priority claim involving Application No. 09/318,500, filed May 25, 1999, now U.S. Patent 6,232,401, however, is in error as it indicates that the above-referenced application, serial No. 10/644,389 ("the '389 application"), is a continuation-in-part of Application No. 09/318,500. The '389 application also erroneously sets forth the relationship of U.S. Provisional Application Nos. 60/182,396 and 60/215,548. The '389 application is not a continuation-in-part of Application No. 09/318,500, it is instead a continuation of 10/388,177, which is a continuation of Application No. 09/756,573, which is, *inter alia*, a continuation-in-part of Application No. 09/318,500, which itself is, *inter alia*, a divisional of Application No. 08/802,130. Application No. 09/756,573 also claims the benefit of the recited provisional applications.

According to 37 C.F.R. § 1.78(a)(2)(ii), the reference to the prior-filed application was to be submitted during the pendency of the later-filed application and within the later of four months from the actual filing date of this application or sixteen (16) months from the filing date of the prior-filed application. As this application was filed on August 19, 2003, and the '500 application was filed on May 25, 1999, the later of four months from the filing date and sixteen months from the filing date of the prior-filed application is four months from the filing date, namely, December 19, 2003.

---

<sup>1</sup> While the filing date of Application No. 10/302,177 is recited in error as November 21, 2001, not 2002, the reference required under 35 U.S.C. § 120 and 37 C.F.R. § 1.78 to claim the benefit of priority requires identification by application number and by the relationship between the applications. 35 U.S.C. § 120; 37 C.F.R. §§ 1.14 and 1.78. This requirement was met despite the incorrect recitation of the year in which the '177 application was filed.

### **Verified Statement**

Applicants hereby state that the entire delay between December 19, 2003, and the date of this petition was unintentional, in accordance with 37 C.F.R. § 1.78(a)(3)(iii).

Applicants first learned that there might be an issue of ambiguity in the recited priority claim involving Application No. 09/318,500 in connection with a pending interference, Patent Interference No. 105,555 (MPT), declared July 10, 2007, and included in its list of proposed motions a “motion with petition to the Board to accept an unintentionally delayed claim for the benefit of a prior-filed application under 37 C.F.R. § 1.78(a)(3), with proposed amendment to involved application no. 10/644,389 to properly claim benefit of priority to application no. 09/318,500 or other appropriate relief.” Applicants sought authorization for a motion, instead of petitioning the PTO immediately, as they understood Applicants must seek relief “in the form of a motion” under the Board’s Standing Order. The motion was authorized by Paper 22, filed August 29, 2007.

### **Required Reference**

Applicants provide herewith an amendment to the specification of the above-referenced application to insert the reference required by 35 U.S.C. § 120 and 37 C.F.R. § 1.78(a)(2)(i), specifying the prior-filed application by its series code, serial number, and indicating the relationship between these applications. Applicants further correct the error in the filing date of Application No. 10/302,177. As amended, the first paragraph of the application entitled “Cross-Reference to Related Applications” will read:

This application is a continuation of U.S. Patent Application No. 10/302,177, filed November 21, 2002; which is a continuation of U.S. Patent Application No. 09/756,573, filed January 8, 2001, now U.S. Patent No. 6,509,418 B1; which is a continuation-in-part of U.S. Patent Application No. 08/928,972, filed September 12, 1997, now U.S. Patent No. 6,172,179;

which is a continuation-in-part of U.S. Patent Application 08/802,130, filed on February 19, 1997, now U.S. Patent No. 5,912,319; said U.S. Patent Application No. 09/756,573 is also a continuation-in-part of U.S. Patent Application No. 09/318,500, filed May 25, 1999, now U.S. Patent No. 6,232,401; which is a division of said U.S. Patent Application No. 08/802,130, filed on February 19, 1997, now U.S. Patent No. 5,912,319; and said U.S. Patent Application No. 09/756,573 claims the benefit of U.S. Provisional Application No. 60/182,396, filed February 14, 2000, and U.S. Provisional Application No. 60/215,548, filed June 30, 2000, all of which are incorporated herein by reference.

**Petition Fee under 37 C.F.R. § 1.17(t)**

Applicants hereby authorize the PTO to charge the petition fee under 37 C.F.R. § 1.17(t) of \$1,410.00 to Deposit Account No. 06-0916.

**Conclusion**

As the requirements under 37 C.F.R. § 1.78(a)(3) for a petition to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120 have been met, Applicants respectfully request that this petition be granted such that the Office recognizes this application's priority claim as set forth above. Please grant any extensions of time required to enter this petition, and should any additional fees be required for the consideration or acceptance of this petition, please charge them to Deposit Account No. 06-0916.

Respectfully submitted,

Date: November 29, 2007

By: \_\_\_\_\_

*Wesley Derrick*

Wesley Derrick  
Reg. No. 46,659  
650-849-6732

**APPENDIX 4**

**PATENT**  
Customer No. 22,852  
Attorney Docket No. 08303.0042-06000

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of: )  
)  
Jonathan D. ZOOK *et al.* ) ATTN: Office of Petitions  
)  
Application No.: 10/644,389 ) Group Art Unit: 1711  
)  
Filed: August 19, 2003 ) Examiner: Duc TRUONG  
)  
For: SEALANTS AND POTTING )  
FORMULATIONS INCLUDING )  
MERCAPTO-TERMINATED )  
POLYMERS PRODUCED BY THE )  
REACTION OF A POLYTHIOL )  
AND POLYVINYL ETHER )  
MONOMER )

**Mail Stop Petition**  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

**AMENDMENT**

In connection with the concurrently submitted petition under 37 C.F.R. § 1.78(a)(3) to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120, please amend this application as follows:

**Amendments to the Specification** is presented at page 2 of this paper.

**Remarks** follow the amendment section of this paper at page 3.

**AMENDMENT TO THE SPECIFICATION**

Please amend the first paragraph of this application, entitled "Cross-Reference to Related Applications," as follows:

This application is a continuation of U.S. Patent Application No. 10/302,177, filed November 21, ~~2001~~ 2002; which is a continuation of U.S. Patent Application No. 09/756,573, filed January 8, 2001, now U.S. Patent No. 6,509,418 B1; which is a continuation-in-part of U.S. Patent Application No. 08/928,972, filed September 12, 1997, now U.S. Patent No. 6,172,179; which is a continuation-in-part of U.S. Patent Application 08/802,130, filed on February 19, 1997, now U.S. Patent No. 5,912,319; ~~and~~ said U.S. Patent Application No. 09/756,573 is also a continuation-in-part of U.S. Patent Application No. 09/318,500, filed May 25, 1999, now U.S. Patent No. 6,232,401; which is a division of said U.S. Patent Application No. 08/802,130, filed on February 19, 1997, now U.S. Patent No. 5,912,319; and said U.S. Patent Application No. 09/756,573 claims the benefit of U.S. Provisional Application No. 60/182,396, filed February 14, 2000, and U.S. Provisional Application No. 60/215,548, filed June 30, 2000, all of which are incorporated herein by reference.

**REMARKS**

This amendment is submitted concurrently with a petition under 37 C.F.R. § 1.78(a)(3) to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120. This submission satisfies the requirement under 37 C.F.R. § 1.78(a)(3)(i) to amend the specification to insert a reference to the application for which the benefit of priority is claimed. Applicants respectfully request that the Office recognizes this application's priority claim as set forth above in the directed amendment.

If there is any fee due in connection with the submission of this Amendment, please charge the fee to Deposit Account No. 06-0916. Please grant any extensions of time required to enter this Amendment and charge any additional required fees to our Deposit Account No. 06-0916.

Respectfully submitted,

Date: November 29, 2007

By: Wesley Derrick  
Wesley Derrick  
Reg. No. 46,659  
650-849-6732

APPENDIX 5

PATENT  
Customer No. 22,852  
Attorney Docket No. 08303.0042-05000

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: )  
)  
Jonathan D. ZOOK *et al.* ) ATTN: Office of Petitions  
)  
Application No.: 10/302,177 ) Group Art Unit: 1711  
)  
Filed: November 21, 2002 ) Examiner: Duc TRUONG  
)  
For: SEALANTS AND POTTING )  
FORMULATIONS INCLUDING )  
MERCAPTO-TERMINATED )  
POLYMERS PRODUCED BY THE )  
REACTION OF A POLYTHIOL )  
AND POLYVINYL ETHER )  
MONOMER )

**Mail Stop Petition**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

**PETITION UNDER 37 C.F.R. § 1.78(a)(3) TO ACCEPT AN  
UNINTENTIONALLY DELAYED CLAIM FOR DOMESTIC PRIORITY**

In a pending interference, *Zook v. Zook*, Patent Interference No. 105,555 (MPT), it has been noted that an alternative priority benefit chain, not the basis of priority in the interference, appears to include some errors. Applicants hereby submit this petition under 37 C.F.R. § 1.78(a)(3) to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120.

The above-referenced application properly claims priority under 35 U.S.C. § 120 to a benefit chain, namely, to Application No. 09/756,573, filed on January 8, 2001, now U.S. Patent 6,509,418, which is a continuation-in-part of Application No. 08/928,972, filed September 12,

1997, now U.S. Patent 6,172,179, which is a continuation-in-part of Application No. 08/802,130, filed February 19, 1997, now U.S. Patent 5,912,319.

Application No. 10/302,177's priority claim involving Application No. 09/318,500, filed May 25, 1999, now U.S. Patent 6,232,401, however, is ambiguous and may allow one to mistakenly conclude that Application No. 10/302,177, is a continuation-in-part of Application No. 09/318,500. This is incorrect; Application No. 10/388,177 is instead a continuation of Application No. 09/756,573, which is, *inter alia*, a continuation-in-part of Application No. 09/318,500, which itself is, *inter alia*, a divisional of Application No. 08/802,130. Similarly, Application No. 09/756,573 also claims the benefit of the recited provisional applications.

According to 37 C.F.R. § 1.78(a)(2)(ii), the reference to the prior-filed application was to be submitted during the pendency of the later-filed application and within the later of four months from the actual filing date of this application or sixteen (16) months from the filing date of the prior-filed application. As this application was filed on November 21, 2002, and the '500 application was filed on May 25, 1999, the later of four months from the filing date and sixteen months from the filing date of the prior-filed application is four months from the filing date, namely, March 21, 2003.

#### **Verified Statement**

Applicants hereby state that the entire delay between March 21, 2003, and the date of this petition was unintentional, in accordance with 37 C.F.R. § 1.78(a)(3)(iii).

Applicants first learned that there might be an issue of ambiguity in the recited priority claim to Application No. 09/318,500 in connection with a pending interference, Patent Interference No. 105,555 (MPT), declared July 10, 2007, and included in its list of proposed motions a "motion with petition to the Board under 37 C.F.R. §§ 1.78(a)(3), 1.137(b) and/or 1.182 to enter an amendment to application no. 10/302,177 indicating priority information for

claim priority to application no. 09/318,500.” Applicants sought authorization for a motion, instead of petitioning the PTO immediately, as they understood Applicants must seek relief “in the form of a motion” under the Board’s Standing Order. The motion was authorized by Paper 22, filed August 29, 2007.

#### **Required Reference**

Applicants provide herewith an amendment to the specification of the above-referenced application to insert the reference required by 35 U.S.C. § 120 and 37 C.F.R. § 1.78(a)(2)(i), specifying the prior-filed application by its series code, serial number, and filing date and indicating the relationship between these applications. As amended, the first paragraph of the application entitled “Cross-Reference to Related Applications” will read:

This application is a continuation of U.S. Patent Application No. 09/756,573, filed January 8, 2001, which is a continuation-in-part of U.S. Patent Application No. 08/928,972, filed September 12, 1997, now U.S. Patent No. 6,172,179, which is a continuation-in-part of U.S. Patent Application No. 08/802,130, filed February 19, 1997, now U.S. Patent No. 5,912,319; said U.S. Patent Application No. 09/756,573 is also a continuation-in-part of U.S. Patent Application No. 09/318,500, filed May 25, 1999, which is a division of said U.S. Patent Application No. 08/802,130, now U.S. Patent No. 5,912,319; and said U.S. Patent Application No. 09/756,573 claims the benefit of U.S. Provisional Application No. 60/182,396, filed February 14, 2000, and U.S. Provisional Application No. 60/215,548, filed June 30, 2000, all of which are incorporated herein by reference.

#### **Petition Fee under 37 C.F.R. § 1.17(t)**

Applicants hereby authorize the PTO to charge the petition fee under 37 C.F.R. § 1.17(t) of \$1,410.00 to Deposit Account No. 06-0916.

**Conclusion**

As the requirements under 37 C.F.R. § 1.78(a)(3) for a petition to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120 have been met, Applicants respectfully request that this petition be granted such that the Office recognizes this application's priority claim as set forth above in the directed amendment.

Please grant any extensions of time required to enter this petition, and should any additional fees be required for the consideration or acceptance of this petition, please charge them to Deposit Account No. 06-0916.

Respectfully submitted,

Date: November 29, 2007

By: Wesley Derrick  
Wesley Derrick  
Reg. No. 46,659  
650-849-6732

**APPENDIX 6**

**PATENT**  
Customer No. 22,852  
Attorney Docket No. 08303.0042-05000

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of: )  
)  
Jonathan D. ZOOK *et al.* ) ATTN: Office of Petitions  
)  
Application No.: 10/302,177 ) Group Art Unit: 1711  
)  
Filed: November 21, 2002 ) Examiner: Duc TRUONG  
)  
For: SEALANTS AND POTTING )  
FORMULATIONS INCLUDING )  
MERCAPTO-TERMINATED )  
POLYMERS PRODUCED BY THE )  
REACTION OF A POLYTHIOL )  
AND POLYVINYL ETHER )  
MONOMER )

**Mail Stop Petition**  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

**AMENDMENT**

In connection with the concurrently submitted petition under 37 C.F.R. § 1.78(a)(3) to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120, please amend this application as follows:

**Amendments to the Specification** is presented at page 2 of this paper.

**Remarks** follow the amendment section of this paper at page 3.

**AMENDMENT TO THE SPECIFICATION**

Please amend the first paragraph of this application, entitled "Cross-Reference to Related Applications," as follows:

This application is a continuation of U.S. Patent Application No. 09/756,573, filed January 8, 2001, which is a continuation-in-part of U.S. Patent Application No. 08/928,972, filed September 12, 1997, now U.S. Patent No. 6,172,179, which is a continuation-in-part of U.S. Patent Application No. 08/802,130, filed February 19, 1997, now U.S. Patent No. 5,912,319; ~~and~~ said U.S. Patent Application No. 09/756,573 is also a continuation-in-part of U.S. Patent Application No. 09/318,500, filed May 25, 1999, which is a division of said U.S. Patent Application No. 08/802,130, now U.S. Patent No. 5,912,319; and said U.S. Patent Application No. 09/756,573 claims the benefit of U.S. Provisional Application No. 60/182,396, filed February 14, 2000, and U.S. Provisional Application No. 60/215,548, filed June 30, 2000, all of which are incorporated herein by reference.

**REMARKS**

This amendment is submitted concurrently with a petition under 37 C.F.R. § 1.78(a)(3) to accept an unintentionally delayed claim for priority under 35 U.S.C. § 120. This submission satisfies the requirement under 37 C.F.R. § 1.78(a)(3)(i) to amend the specification to insert a reference to the application for which the benefit of priority is claimed. Applicants respectfully request that the Office recognizes this application's priority claim as set forth above in the directed amendment.

If there is any fee due in connection with the submission of this Amendment, please charge the fee to Deposit Account No. 06-0916. Please grant any extensions of time required to enter this Amendment and charge any additional required fees to our Deposit Account No. 06-0916.

Respectfully submitted,

Date: November 29, 2007

By: Wesley Derrick  
Wesley Derrick  
Reg. No. 46,659  
650-849-6732

**CERTIFICATE OF SERVICE**

I hereby certify that a copy of the foregoing, **SENIOR PARTY ZOOK (PRC)**  
**MOTION 2**, was served on the Junior Party Zook through its attorneys of record on this the 30th  
day of November, 2007 as follows:

**VIA ELECTRONIC MAIL:**

Mark D. Schneider, Esq.  
Douglas W. Sprinkle Esq.  
Avery N. Goldstein, Esq.  
Grifford, Krass, Sprinkle, Anderson & Citkowski, P.C.  
Suite 330  
2701 Troy Center Drive  
Post Office Box 7021  
Troy, Michigan 48007  
Telephone: 248.647.6000  
Facsimile: 248.647.5210  
e-mail: litigation@patlaw.com

Respectfully submitted,

By: /Jerry D. Voight/  
Jerry D. Voight  
Registration No. 23,020

Attorney of Record for Senior Party Zook

Filed on behalf of: Senior Party Jonathan D. Zook

By: Jerry D. Voight  
Wesley B. Derrick  
FINNEGAN, HENDERSON, FARABOW,  
GARRETT & DUNNER, L.L.P.  
3300 Hillview Avenue  
Palo Alto, California 94304  
Telephone: 650.849.6600  
Facsimile: 650.849.6666

UNITED STATES PATENT AND TRADEMARK OFFICE

---

BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

---

JONATHAN D. **ZOOK**, DAVID W. JORDAN, DEAN M. WILLARD,  
GEORGE JONES, AND MICHAEL COSMAN,  
Junior Party ("PBT")  
(U.S. Patents 6,525,168 and 6,723,827),

v.

JONATHAN D. **ZOOK**, SUSAN E. DEMOSS,  
DAVID W. JORDAN AND CHANDRA B. RAO  
Senior Party ("PRC")  
(U.S. Application 10/644,389).

---

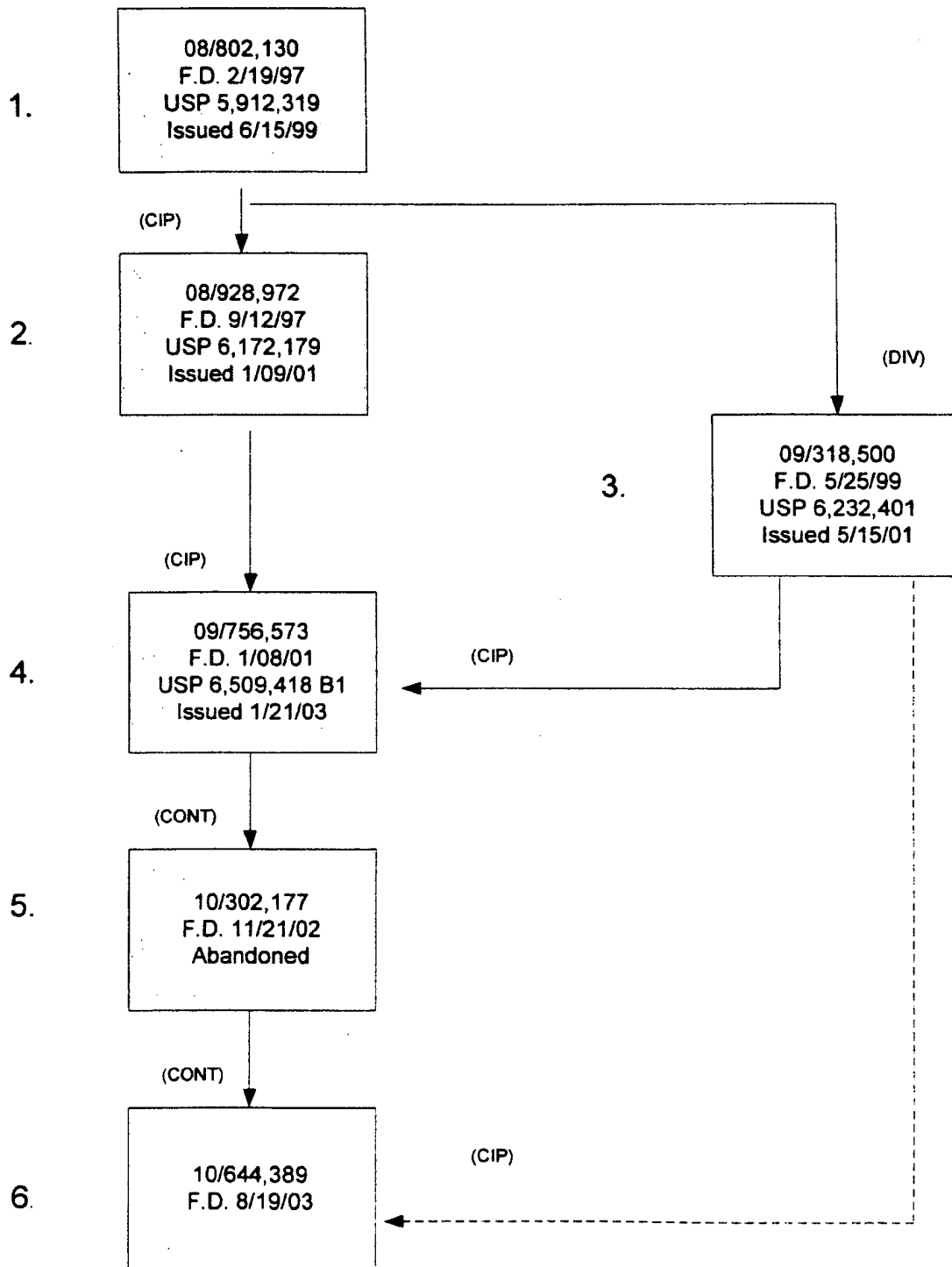
Patent Interference No. 105,555 (MPT)  
(Technology Center 1700)

---

**ZOOK EXHIBIT 1003**

**ZOOK EXHIBIT 1003**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

SENIOR PARTY ZOOK (PRC) INVOLVED APPLICATION  
FAMILY TREE



16669 U.S. PTO  
08/19/03

PATENT  
Customer Number 22,852  
Attorney Docket No. 08303.0042-06

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

**Mail Stop Patent Application**  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

22212 U.S. PTO  
10/644389  
08/19/03

Prior Application Art Unit: 1711 Prior Application Examiner: Duc Truong

SIR: This is a request for filing a Continuation Application under 37 C.F.R. § 1.53(b) of pending prior Application No. 10/302,177, filed November 21, 2001, of Jonathan D. Zook, Susan E. DeMoss, David W. Jordan, Chandra B. Rao, Hakam Singh, and Ahmed Sharaby, for SEALANTS AND POTTING FORMULATIONS INCLUDING MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER.

1. ☒ Enclosed is a complete copy of the prior application including the oath or Declaration and drawings, if any, as originally filed. On information and belief, I hereby verify that the attached papers are a true copy of prior Application No. 10/302,177, and the Declaration and Power of Attorney as originally filed on November 21, 2001, which are incorporated herein by reference.
2. ☐ Enclosed is a substitute specification under 37 C.F.R. § 1.125. The undersigned hereby verifies that no new matter is added in this substitute specification.
3. ☐ Enclosed is a Request for Non-Publication of Application and Certification Under 35 U.S.C. § 122(b)(2)(B)(i).
4. ☒ Cancel Claims 1-21.
5. ☒ A Preliminary Amendment is enclosed.
6. ☒ The filing fee is calculated on the basis of the claims existing in the prior application as amended at 4 and 5 above.

FINNEGAN  
HENDERSON  
FARABOW  
GARRETT &  
DUNNER LLP

1300 I Street, NW  
Washington, DC 20005  
202.408.4000  
Fax 202.408.4400  
www.finnegan.com

**ZOOK EXHIBIT 1006**  
Zook v. Zook  
Interference No. 105,555 (MPT)

Basic Application Filing Fee					\$750	\$ \$750.00
	Number of Claims		Basic	Extra Claims		
Total Claims	17	-	20	0	x \$18	
Independent Claims	2	-	3	0	x \$84	
<input type="checkbox"/> Presentation of Multiple Dep. Claim(s)					+\$280	
Subtotal						\$ 750.00
Reduction by 1/2 if small entity						-
TOTAL APPLICATION FILING FEE						\$ 750.00

7. ☒ Please charge \$750.00 to Deposit Account No. 06-0916, to cover the filing fee.
8. ☒ The Commissioner is hereby authorized to charge any fees which may be required including fees due under 37 C.F.R. § 1.16 and any other fees due under 37 C.F.R. § 1.17, or credit any overpayment during the pendency of this application to Deposit Account No. 06-0916.
9. ☒ Amend the specification by replacing the first paragraph on page 1, under the sub-heading Cross-Reference to Related Applications, with the following new paragraph:

This application is a continuation of U.S. Patent Application 10/302,177, filed November 21, 2001; which is a continuation of U.S. Patent Application No. 09/756,573, filed January 8, 2001, now U.S. Patent No. 6,509,418 B1; which is a continuation-in-part of U.S. Patent Application 08/928,972, filed September 12, 1997, now U.S. Patent No. 6,172,179; which is a continuation-in-part of U.S. Patent Application 08/802,130, filed on February 19, 1997, now U.S. Patent No. 5,912,319; and also a continuation-in-part of U.S. Patent Application No. 09/318,500, filed May 25, 1999, now U.S. Patent No. 6,232,401; which is a division of U.S. Patent Application No. 08/802,130, filed on February 19, 1997, now U.S. Patent No. 5,912,319; and claims the benefit of U.S. Provisional Application No. 60/182,396, filed February 14, 2000, and U.S. Provisional Application No. 60/215,548, filed June 30, 2000, all of which are incorporated herein by reference.

10. ☐ New acceptable drawings are enclosed.

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11. ☒ The prior application is assigned of record to: PRC-DeSoto International, Inc., 5430 San Fernando Road, Glendale, California 91209.
12. ☐ Priority of Application No. [Text], filed on [Text] in [Country] is claimed under 35 U.S.C. § 119. A certified copy
- ☐ is enclosed or ☐ is on file in the prior application.
13. ☐ Small entity status is appropriate and applies to this application.
14. ☒ The power of attorney in the prior application is to at least one of the following: FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P., Douglas B. Henderson, Reg. No. 20,291; Ford F. Farabow, Jr., Reg. No. 20,630; Arthur S. Garrett, Reg. No. 20,338; Donald R. Dunner, Reg. No. 19,073; Brian G. Brunsvold, Reg. No. 22,593; Tipton D. Jennings, IV, Reg. No. 20,645; Jerry D. Voight, Reg. No. 23,020; Laurence R. Hefter, Reg. No. 20,827; Kenneth E. Payne, Reg. No. 23,098; Herbert H. Mintz, Reg. No. 26,691; C. Larry O'Rourke, Reg. No. 26,014; Albert J. Santorelli, Reg. No. 22,610; Michael C. Elmer, Reg. No. 25,857; Richard H. Smith, Reg. No. 20,609; Stephen L. Peterson, Reg. No. 26,325; John M. Romary, Reg. No. 26,331; Bruce C. Zotter, Reg. No. 27,680; Dennis P. O'Reilly, Reg. No. 27,932; Allen M. Sokal, Reg. No. 26,695; Robert D. Bajefsky, Reg. No. 25,387; Richard L. Stroup, Reg. No. 28,478; David W. Hill, Reg. No. 28,220; Thomas L. Irving, Reg. No. 28,619; Charles E. Lipsey, Reg. No. 28,165; Thomas W. Winland, Reg. No. 27,605; Basil J. Lewis, Reg. No. 28,818; Martin I. Fuchs, Reg. No. 28,508; E. Robert Yoches, Reg. No. 30,120; Barry W. Graham, Reg. No. 29,924; Susan Haberman Griffen, Reg. No. 30,907; Richard B. Racine, Reg. No. 30,415; Thomas H. Jenkins, Reg. No. 30,857; Robert E. Converse, Jr., Reg. No. 27,432; Clair X. Mullen, Jr., Reg. No. 20,348; Christopher P. Foley, Reg. No. 31,354; John C. Paul, Reg. No. 30,413; David M. Kelly, Reg. No. 30,953; Kenneth J. Meyers, Reg. No. 25,146; Carol P. Einaudi, Reg. No. 32,220; Walter Y. Boyd, Jr., Reg. No. 31,738; Steven M. Anzalone, Reg. No. 32,095; Jean B. Fordis, Reg. No. 32,984; Roger D. Taylor, Reg. No. 28,992; Barbara C. McCurdy, Reg. No. 32,120; James K. Hammond, Reg. No. 31,964; Richard V. Burgujian, Reg. No. 31,744; J. Michael Jakes, Reg. No. 32,824; Thomas W. Banks, Reg. No. 32,719; Christopher P. Isaac, Reg. No. 32,616; Bryan C. Diner, Reg. No. 32,409; M. Paul Barker, Reg. No. 32,013; Andrew Chanho Sonu, Reg. No. 33,457; David S. Forman, Reg. No. 33,694; Vincent P. Kovalick, Reg. No. 32,867; James W. Edmondson, Reg. No. 33,871; Michael R. McGurk, Reg. No. 32,045; Joann M. Neth, Reg. No. 36,363; Gerson S. Panitch, Reg. No. 33,751; Cheri M. Taylor, Reg. No. 33,216; Charles E. Van Horn, Reg. No. 40,266; Linda A. Wadler, Reg. No. 33,218; Jeffrey A. Berkowitz, Reg. No. 36,743; Michael R. Kelly, Reg. No. 33, 921; James B. Monroe, Reg. No. 33,971; Doris Johnson Hines, Reg. No. 34,629; Lori Ann Johnson, Reg. No. 34,498; R. Bruce Bower, Reg. No. 37,099; John

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Rissman, Reg. No. 33,764; Therese A. Hendricks, Reg. No. 30,389; Leslie I. Bookoff, Reg. No. 38,084; Michele C. Bosch, Reg. No. 40,524; Michael J. Flibbert, Reg. No. 33,234; Scott A. Herbst, Reg. No. 35,189; Leslie A. McDonell, Reg. No. 34,872; Thalia V. Warnement, Reg. No. 39,064; Ronald A. Bleeker, Reg. No. 27,773; Kathleen A. Daley, Reg. No. 36,116; C. Gregory Gramenopoulos, Reg. No. 36,532; Anthony M. Gutowski, Reg. No. 38,742; Yitai Hu, Reg. No. 40,653; Lionel M. Lavenue, Reg. No. 46,859; Christine E. Lehman, Reg. No. 38,535; Patrick J. Coyne, Reg. No. 31,821; and William R. Lambert, Reg. No. 44,857.

15. ☐ The power appears in the original declaration of the prior application.
16. ☐ Since the power does not appear in the original declaration, a copy of the power in the prior application is enclosed.
17. ☒ Please address all correspondence to FINNEGAN, HENDERSON, FARABOW, GARRETT and DUNNER, L.L.P., 1300 I Street, N.W., Washington, D.C. 20005-3315, Customer Number 22,852.
18. ☐ Recognize as associate attorney [Name, Address & Reg. No.]
19. ☐ Also enclosed is \_\_\_\_\_

**PETITION FOR EXTENSION.** If any extension of time is necessary for the filing of this application, including any extension in parent Application No. 10/302,177, filed November 21, 2001, for the purpose of maintaining copendency between the parent application and this application, and such extension has not otherwise been requested, such an extension is hereby requested, and the Commissioner is authorized to charge necessary fees for such an extension to Deposit Account No. 06-0916.

FINNEGAN, HENDERSON, FARABOW,  
GARRETT & DUNNER, L.L.P.

Dated: August 19, 2003

By: \_\_\_\_\_

William R. Lambert  
Reg. No. 44,857

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**CERTIFICATE OF EXPRESS MAILING**

Express Mail Label No.: EV 351294430 US

I hereby certify that this correspondence is being deposited with the United States Postal Services "Express Mail Post Office to Addressee" service under 37 C.F.R. § 1.10, addressed to: Mail Stop Patent Application, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the date below.

Date: August 19, 2003

Signed: \_\_\_\_\_

Linda Phillips  
Linda Phillips

11/21/02  
JC031 U.S. PTO

11-25-02

PATENT  
Customer Number 22,852  
Attorney Docket No. 8303.0042-05

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
COMMISSIONER FOR PATENTS  
Washington, D.C. 20231

Prior Application Art Unit: 1711 Prior Application Examiner: Duc Truong

SIR: This is a request for filing a **Continuation Application** under 37 C.F.R. § 1.53(b) of pending prior Application No. 09/756,573 filed January 8, 2001, of Jonathan D. Zook, Susan E. DeMoss, David W. Jordan, Chandra B. Rao, Hakam Singh, and Ahmed Sharaby for SEALANTS AND POTTING FORMULATIONS INCLUDING MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER.

1. ☒ Enclosed is a complete copy of the prior application including the oath or Declaration and drawings, if any, as originally filed. I hereby verify that the attached papers are true copies of prior Application No. 09/756,573 as originally filed on January 8, 2001, and of the Declaration filed on June 13, 2001, which is incorporated herein by reference.
2. ☐ Enclosed is a substitute specification under 37 C.F.R. § 1.125.
3. ☐ Enclosed is a Request for Non-Publication of Application and Certification Under 35 U.S.C. § 122(b)(2)(B)(i).
4. ☒ Cancel Claims 1-21.
5. ☒ A Preliminary Amendment is enclosed.
6. ☒ The filing fee is calculated on the basis of the claims existing in the prior application as amended at 4 and 5 above:

Basic Application Filing Fee					\$740	\$ \$740.00
	Number of Claims		Basic	Extra Claims		
Total Claims	19	-	20	0	x \$18	
Independent Claims	3	-	3	0	x \$84	
<input type="checkbox"/> Presentation of Multiple Dep. Claim(s)					+\$280	
Subtotal						\$ 740.00
Reduction by 1/2 if small entity						-
TOTAL APPLICATION FILING FEE						\$ 740.00

ZOOK EXHIBIT 1007  
Zook v. Zook  
Interference No. 105,555 (MPT)

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7. ☒ Please charge \$740.00 to Deposit Account 06-0916 to cover the filing fee.
8. ☒ The Commissioner is hereby authorized to charge any fees which may be required including fees due under 37 C.F.R. § 1.16 and any other fees due under 37 C.F.R. § 1.17, or credit any overpayment during the pendency of this application to Deposit Account No. 06-0916.
9. ☒ Amend the specification by inserting before the first line, the following new paragraph:  
  
This is a continuation of U.S. Patent Application No. 09/756,573, filed January 8, 2001, which is a continuation-in-part of U.S. Patent Application No. 08/928,972, filed September 12, 1997, which is a continuation-in-part of now U.S. Patent No. 5,912,319; and also a continuation-in-part of U.S. Patent Application No. 09/318,500, filed May 25, 1999, which is a division of now U.S. Patent No. 5,912,319; and claims the benefit of U.S. Provisional Application No. 60/182,396, filed February 14, 2000, and U.S. Provisional Application No. 60/215,548, filed June 30, 2000, all of which are incorporated herein by reference.
10. ☐ New formal drawings are enclosed.
11. ☒ The prior application is assigned of record to: PRC-DeSoto International, Inc., 5430 San Fernando Road, Glendale, California 91209.
12. ☐ Priority of Application No. [Text], filed on [Text] in [Country] is claimed under 35 U.S.C. § 119. A certified copy  
  
☐ is enclosed or ☐ is on file in the prior application.
13. ☐ Small entity status is appropriate and applies to this application.
14. ☒ The power of attorney in the prior application is to at least one of the following: FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P., Douglas B. Henderson, Reg. No. 20,291; Ford F. Farabow, Jr., Reg. No. 20,630; Arthur S. Garrett, Reg. No. 20,338; Donald R. Dunner, Reg. No. 19,073; Brian G. Brunsvold, Reg. No. 22,593; Tipton D. Jennings, IV, Reg. No. 20,645; Jerry D. Voight, Reg. No. 23,020; Laurence R. Hefter, Reg. No. 20,827; Kenneth E. Payne, Reg. No. 23,098; Herbert H. Mintz, Reg. No. 26,691; C. Larry O'Rourke, Reg. No. 26,014; Albert J. Santorelli, Reg. No. 22,610; Michael C. Elmer, Reg.

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15. ☐ The power appears in the original declaration of the prior application.
16. ☐ Since the power does not appear in the original declaration, a copy of the power in the prior application is enclosed.

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17. ☒ Please address all correspondence to FINNEGAN, HENDERSON, FARABOW, GARRETT and DUNNER, L.L.P., 1300 I Street, N.W., Washington, D.C. 20005-3315, **Customer Number 22,852.**
18. ☒ Recognize as associate attorney William R. Lambert, Reg. No. 44,367, of Finnegan, Henderson, Farabow, Garrett & Dunner, L.L.P.
19. ☐ Also enclosed is \_\_\_\_\_

**PETITION FOR EXTENSION.** If any extension of time is necessary for the filing of this application, including any extension in parent Application No. 09/756,573, filed January 8, 2002, for the purpose of maintaining copendency between the parent application and this application, and such extension has not otherwise been requested, such an extension is hereby requested, and the Commissioner is authorized to charge necessary fees for such an extension to Deposit Account No. 06-0916. A duplicate copy of this paper is enclosed for use in charging the deposit account.

FINNEGAN, HENDERSON, FARABOW,  
GARRETT & DUNNER, L.L.P.

Dated: November 21, 2002

By: 

Phil M. Makrogiannis  
Reg. No. 47,766

CERTIFICATE OF EXPRESS MAILING

Express Mail Label No.: EL 746265368 US

I hereby certify that this correspondence is being deposited with the United States Postal Services "Express Mail Post Office to Addressee" service under 37 C.F.R. § 1.10, addressed to Box Patent Applications, Commissioner for Patents, Washington, D.C. 20231, on the date below.

Date: November 21, 2002

Signed: 

Linda Phillips

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HENDERSON 650 849 6666 TO

213408449

P. 86

Customer Number 22,852  
Attorney Docket No. 6303.0042-04

## DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: **SEALANTS AND POTTING FORMULATIONS MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER**, the specification of which ☐ is attached and/or ☒ was filed on January 8, 2001, as United States Application Serial No. 09/758,573 or PCT International Application No. \_\_\_\_\_ and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(e)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate or § 365(a) of any PCT International application(s) designating at least one country other than the United States, listed below and have also identified below, any foreign application(s) for patent or inventor's certificate, or any PCT International application(s) having a filing date before that of the application(s) of which priority is claimed:

Country	Application Number	Date of Filing	Priority Claimed Under 35 U.S.C. 119
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

Application Number	Date of Filing
60/182,396	February 14, 2000
60/215,548	June 30, 2000

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s) or § 365(c) of any PCT International application(s) designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application(s) in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application(s) and the national or PCT International filing date of this application:

Application Number	Date of Filing	Status (Patented, Pending, Abandoned)
09/316,500	May 25, 1999	Pending
08/826,972	September 12, 1997	Patented
08/802,130	February 19, 1997	Patented

I hereby appoint the following attorney and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: **FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**, CUSTOMER NUMBER 22,852, Douglas B. Henderson, Reg. No. 20,294; Ford F. Farabow, Jr., Reg. No. 20,630; Arthur S. Garrett, Reg. No. 20,338; Donald R. Dunner, Reg. No. 19,073; Brian G. Brunsvold, Reg. No. 22,593; Tipton D. Jennings, IV, Reg. No. 20,645; Jerry D. Voight, Reg. No. 23,020; Laurence R. Heffer, Reg. No. 20,827; Kenneth E. Payne, Reg. No. 23,088; Herbert H. Mintz, Reg. No. 26,691; C. Larry O'Rourke, Reg. No. 26,014; Albert J. Santorelli, Reg. No. 22,610; Michael C. Elmer, Reg. No. 25,857; Richard H. Smith, Reg. No. 20,609; Stephen L. Peterson, Reg. No. 26,325; John M. Romary, Reg. No. 26,331; Bruce C. Zoller, Reg. No. 27,680; Dennis P. O'Reilly, Reg. No. 27,932; Allen M. Sokal, Reg. No. 26,695; Robert D. Bajelsky, Reg. No. 25,387; Richard L. Stroup, Reg. No. 28,478; David W. Hill, Reg. No. 28,220; Thomas L. Irving, Reg. No. 28,619; Charles E. Lipsey, Reg. No. 28,166; Thomas W. Winland, Reg. No. 27,605; Basil J. Lewis, Reg. No. 28,816; Martin I. Fuchs, Reg. No. 28,508; E. Robert Yoches, Reg. No. 30,120; Barry W. Graham, Reg. No. 29,824; Susan Haberman Griffin, Reg. No. 30,907; Richard B. Racine, Reg. No. 30,415; Thomas H. Jenkins, Reg. No. 30,657; Robert E. Converse, Jr., Reg. No. 27,432; Clark X. Mullen, Jr., Reg. No. 20,348; Christopher P. Foley, Reg. No. 31,364; John C. Paul, Reg. No. 30,413; Roger D. Taylor, Reg. No. 28,992; David M. Kelly, Reg. No. 30,953; Kenneth J. Meyers, Reg. No. 25,146; Carol P. Einsaui, Reg. No. 32,220; Walter Y. Boyd, Jr., Reg. No. 31,738; Steven M. Anzalone, Reg. No. 32,095; Jean B. Fordis, Reg. No. 32,984; Barbara C. McCurdy, Reg. No. 32,120; James K. Hammond, Reg. No. 31,964; Richard V. Burguljan, Reg. No. 31,744; J. Michael Jakes, Reg. No. 32,824; Thomas W. Banks, Reg. No. 32,719; Christopher P. Isaac, Reg. No. 32,616; Bryan C. Diner, Reg. No. 32,408; M. Paul Barker, Reg. No. 32,013; Andrew Chanhon Sonu, Reg. No. 33,457; David S. Forman, Reg. No. 33,694; Vincent P. Kovalick, Reg. No. 32,867; James W. Edmondson, Reg. No. 33,871; Michael R. McGurk, Reg. No. 32,045; Joann M. Nath, Reg. No. 36,363; Gerson S. Panitch, Reg. No. 33,751; Cheryl M. Taylor, Reg. No. 33,216; Charles E. Van Horn, Reg. No. 40,266; Linda A. Wadler, Reg. No. 33,218; Jeffrey A. Berkowitz, Reg. No. 36,743; Michael R. Kelly, Reg. No. 33,921; James B. Monroe, Reg. No. 33,971; Doris Johnson Hines, Reg. No. 34,628; Allen R. Jensen, Reg. No. 28,224; Lori Ann Johnson, Reg. No. 34,498; David A. Manapogizer, Reg. No. 37,540; Robert J. Eichelsburg, Reg. No. 23,967; Phil N. Makrogiannis, Reg. No. P-47,766; and Linda Pincione, Reg. No. 30,474. Please address all correspondence to **FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**, 1300 I Street, N.W., Washington, D.C. 20005, Telephone No. (202) 408-4000.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Listing of Inventors on Page 2 hereof.

January 2000

**FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**

**ZOOK EXHIBIT 1008**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

APR 25 2001 17:20 FR FINNE

HENDERSON 650 849 6666 TO

J13408449

P. 07

Listing of Inventors From Page 1 hereof.

Attorney Docket No. 6303.0042-04

Full Name of First Inventor Jonathan D. Zook	Inventor's Signature <i>Jonathan D. Zook</i>	Date 1-26-01
Residence 20303 High Point Place, Santa Clarita, CA 91351		Citizenship United States of America
Post Office Address Same as above		
Full Name of Second Inventor Susan E. DeMoss	Inventor's Signature	Date
Residence 17420 Van Owen Street, #5, Van Nuys, CA 91406		Citizenship United States of America
Post Office Address Same as above		
Full Name of Third Inventor David W. Jordan	Inventor's Signature	Date
Residence 10960 Des Moines Avenue, Northridge, CA 91326		Citizenship United States of America
Post Office Address Same as above		
Full Name of Fourth Inventor Chandra B. Rao	Inventor's Signature	Date
Residence 25716 North Player Drive, #T-15, Valencia, CA 91355		Citizenship United States of America
Post Office Address Same as above		
Full Name of Fifth Inventor Hakam Singh	Inventor's Signature	Date
Residence 706 Winston Avenue, Bradbury, CA 91010		Citizenship United States of America
Post Office Address Same as above		
Full Name of Sixth Inventor Ahmed Sharaby	Inventor's Signature	Date
Residence 15666 Falconrim, Canyon Country, CA 91351		Citizenship United States of America
Post Office Address Same as above		
Full Name of Seventh Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		
Full Name of Eighth Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		

## DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; I believe I am the original, first, and sole inventor (if only one name is listed below) or an original, first, and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: **SEALANTS AND POTTING FORMULATIONS MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER**, the specification of which ☐ is attached and/or ☒ was filed on January 8, 2001, as United States Application Serial No. 09/756,573 or PCT International Application No. \_\_\_\_\_ and was amended on \_\_\_\_\_ (if applicable).

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08/928,972	September 12, 1997	Patented
08/802,130	February 19, 1997	Patented

I hereby appoint the following attorney and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: **FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**, CUSTOMER NUMBER 22,852, Douglas B. Henderson, Reg. No. 20,291; Ford F. Farabow, Jr., Reg. No. 20,630; Arthur S. Garrett, Reg. No. 20,338; Donald R. Dunner, Reg. No. 19,073; Brian G. Brunsvold, Reg. No. 22,593; Tipton D. Jennings, IV, Reg. No. 20,645; Jerry D. Voight, Reg. No. 23,020; Laurence R. Heffer, Reg. No. 20,827; Kenneth E. Payne, Reg. No. 23,098; Herbert H. Mintz, Reg. No. 26,691; C. Larry O'Rourke, Reg. No. 26,014; Albert J. Santorelli, Reg. No. 22,610; Michael C. Elmer, Reg. No. 25,857; Richard H. Smith, Reg. No. 20,609; Stephen L. Peterson, Reg. No. 26,325; John M. Romary, Reg. No. 26,331; Bruce C. Zotter, Reg. No. 27,680; Dennis P. O'Reilly, Reg. No. 27,932; Allen M. Sokal, Reg. No. 26,695; Robert D. Bajefsky, Reg. No. 25,387; Richard L. Stroup, Reg. No. 28,478; David W. Hill, Reg. No. 28,220; Thomas L. Irving, Reg. No. 28,619; Charles E. Lipsey, Reg. No. 28,165; Thomas W. Winland, Reg. No. 27,605; Basil J. Lewis, Reg. No. 28,818; Martin I. Fuchs, Reg. No. 28,508; E. Robert Yoches, Reg. No. 30,120; Barry W. Graham, Reg. No. 29,924; Susan Haberman Griffen, Reg. No. 30,907; Richard B. Racine, Reg. No. 30,415; Thomas H. Jenkins, Reg. No. 30,857; Robert E. Converse, Jr., Reg. No. 27,432; Clair X. Mullen, Jr., Reg. No. 20,348; Christopher P. Foley, Reg. No. 31,354; John C. Paul, Reg. No. 30,413; Roger D. Taylor, Reg. No. 28,992; David M. Kelly, Reg. No. 30,953; Kenneth J. Meyers, Reg. No. 25,146; Carl P. Einaudi, Reg. No. 32,220; Walter Y. Boyd, Jr., Reg. No. 31,738; Steven M. Anzalone, Reg. No. 32,095; Jean B. Fordis, Reg. No. 32,984; Barbara C. McCurdy, Reg. No. 32,120; James K. Hammond, Reg. No. 31,964; Richard V. Burgujian, Reg. No. 31,744; J. Michael Jakes, Reg. No. 32,824; Thomas W. Banks, Reg. No. 32,719; Christopher P. Isaac, Reg. No. 32,616; Bryan C. Diner, Reg. No. 32,409; M. Paul Barker, Reg. No. 32,013; Andrew Chanho Sonu, Reg. No. 33,457; David S. Forman, Reg. No. 33,694; Vincent P. Kovalick, Reg. No. 32,867; James W. Edmondson, Reg. No. 33,871; Michael R. McGurk, Reg. No. 32,045; Joann M. Neth, Reg. No. 36,363; Gerson S. Panitch, Reg. No. 33,751; Cheryl M. Taylor, Reg. No. 33,216; Charles E. Van Horn, Reg. No. 40,266; Linda A. Wadler, Reg. No. 33,218; Jeffrey A. Berkowitz, Reg. No. 36,743; Michael R. Kelly, Reg. No. 33,921; James B. Monroe, Reg. No. 33,971; Doris Johnson Hines, Reg. No. 34,629; Allen R. Jensen, Reg. No. 28,224; Lori Ann Johnson, Reg. No. 34,498; David A. Manspeizer, Reg. No. 37,540; Robert J. Eichelburg, Reg. No. 23,057; Phil N. Makrogiannis, Reg. No. P-47,766; and Linda Pingitore, Reg. No. 30,414. Please address all correspondence to **FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.** 1300 I Street, N.W., Washington, D.C. 20005, Telephone No. (202) 408-4000.

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Listing of Inventors on Page 2 hereof.

**FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**

January 2000

Full Name of First Inventor Jonathan D. Zook	Inventor's Signature	Date
Residence 20303 High Point Place, Santa Clarita, CA 91351		Citizenship United States of America
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Full Name of Second Inventor Susan E. DeMoss	Inventor's Signature <i>Susan E. DeMoss</i>	Date 4/20/01
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Post Office Address Same as above		
Full Name of Third Inventor David W. Jordan	Inventor's Signature	Date
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Post Office Address Same as above		
Full Name of Fourth Inventor Chandra B. Rao	Inventor's Signature <i>Chandra B. Rao</i>	Date 4/23/01
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Post Office Address Same as above		
Full Name of Fifth Inventor Hakam Singh	Inventor's Signature <i>Hakam Singh</i>	Date 4/23/01
Residence 706 Winston Avenue, Bradbury, CA 91010		Citizenship United States of America
Post Office Address Same as above		
Full Name of Sixth Inventor Ahmed Sharaby	Inventor's Signature <i>Ahmed Sharaby</i>	Date 4/20/01
Residence 15866 Falconrim, Canyon Country, CA 91351		Citizenship United States of America
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Listing of Inventors on Page 2 hereof.

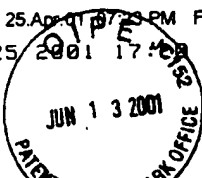
**FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**

January 2000

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Residence 20303 High Point Place, Santa Clarita, CA 91351		Citizenship United States of America
Post Office Address Same as above		
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APR 25 2001 17:03 FR F FINNEGAN HENDERSON 650 849 6 TO 18013408449

P.06

Customer Number 22,852  
Attorney Docket No. 8303.0042-04

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January 2000

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**ZOOK EXHIBIT 1009**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

APR 25 2001 17:20 FR F NEGAN HENDERSON 650 849 667 TO 18013408449

P.07

Listing of Inventors From Page 1 hereof.

Attorney Docket No. 6303.0042-04

Full Name of First Inventor Jonathan D. Zook	Inventor's Signature <i>Jonathan D. Zook</i>	Date 4-26-01
Residence 20303 High Point Place, Santa Clarita, CA 91361		Citizenship United States of America
Post Office Address Same as above		
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60/215,548	June 30, 2000

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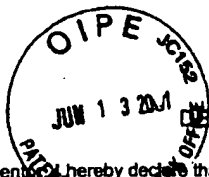
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Listing of Inventors on Page 2 hereof.

January 2000

**FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**

Full Name of First Inventor Jonathan D. Zook	Inventor's Signature	Date
Residence 20303 High Point Place, Santa Clarita, CA 91351		Citizenship United States of America
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Full Name of Second Inventor Susan E. DeMoss	Inventor's Signature <i>Susan E. DeMoss</i>	Date 4/20/01
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Full Name of Fourth Inventor Chandra B. Rao	Inventor's Signature <i>Chandra B. Rao</i>	Date 4/23/01
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Full Name of Sixth Inventor Ahmed Sharaby	Inventor's Signature <i>Ahmed Sharaby</i>	Date 4/25/01
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Post Office Address Same as above		
Full Name of Seventh Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		
Full Name of Eighth Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		



Customer Number 22,852  
Attorney Docket No. 8303.0042-04

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Listing of Inventors on Page 2 hereof.

**FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.**

January 2000

Full Name of First Inventor <b>Jonathan D. Zook</b>	Inventor's Signature	Date
Residence 20303 High Point Place, Santa Clarita, CA 91351		Citizenship United States of America
Post Office Address Same as above		
Full Name of Second Inventor <b>Susan E. DeMoss</b>	Inventor's Signature	Date
Residence 17420 Van Owen Street, #5, Van Nuys, CA 91408		Citizenship United States of America
Post Office Address Same as above		
Full Name of Third Inventor <b>David W. Jordan</b>	Inventor's Signature <i>David W. Jordan</i>	Date 1/25/01
Residence 10960 Des Moines Avenue, Northridge, CA 91326		Citizenship United States of America
Post Office Address Same as above		
Full Name of Fourth Inventor <b>Chandra B. Rao</b>	Inventor's Signature	Date
Residence 25716 North Player Drive, #T-15, Valencia, CA 91355		Citizenship United States of America
Post Office Address Same as above		
Full Name of Fifth Inventor <b>Hakam Singh</b>	Inventor's Signature	Date
Residence 706 Winston Avenue, Bradbury, CA 91010		Citizenship United States of America
Post Office Address Same as above		
Full Name of Sixth Inventor <b>Ahmed Sharaby</b>	Inventor's Signature	Date
Residence 15866 Falconrim, Canyon Country, CA 91351		Citizenship United States of America
Post Office Address Same as above		
Full Name of Seventh Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		
Full Name of Eighth Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		

APR 25 2001 17:20 FR FINNEGAN HENDERSON 650 849 6666 TO 8013408449

P. 06

Customer Number 22,852  
Attorney Docket No. 6303.0042-04

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ZOOK EXHIBIT 1010  
Zook v. Zook  
Interference No. 105,555 (MPT)

APR 25 2001 17:20 FR FINNEGAN HENDERSON 650 849 6666 TO 8013408449

P.06

Customer Number 22,852  
Attorney Docket No. 6303.0042-04

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Listing of inventors on Page 2 hereof.

January 2000

FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.

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Listing of Inventors From Page 1 heretof.

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Full Name of Eighth Inventor	Inventor's Signature	Date
Residence		Citizenship
Post Office Address		

## DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; I believe I am the original, first, and sole inventor (if only one name is listed below) or an original, first, and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: **SEALANTS AND POTTING FORMULATIONS MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER**, the specification of which ☐ is attached and/or ☒ was filed on January 8, 2001, as United States Application Serial No. 09/756,573 or PCT International Application No. \_\_\_\_\_ and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate or § 365(a) of any PCT international application(s) designating at least one country other than the United States, listed below and have also identified below, any foreign application(s) for patent or inventor's certificate, or any PCT international application(s) having a filing date before that of the application(s) of which priority is claimed:

Country	Application Number	Date of Filing	Priority Claimed Under 35 U.S.C. 119
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below:

Application Number	Date of Filing
60/182,398	February 14, 2000
60/215,548	June 30, 2000

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Listing of Inventors on Page 2 hereof.

January 2000

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January 2000

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Full Name of Third Inventor David W. Jordan	Inventor's Signature <i>David Jordan</i>	Date 1/25/01
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(12) **United States Patent**  
Zook et al.

(10) Patent No.: **US 6,509,418 B1**  
(45) Date of Patent: **\*Jan. 21, 2003**

(54) **SEALANTS AND POTTING FORMULATIONS INCLUDING MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER**

(75) Inventors: Jonathan D. Zook, Santa Clarita, CA (US); Susan E. DeMoss, Van Nuys, CA (US); David W. Jordan, Northridge, CA (US); Chandra B. Rao, Valencia, CA (US); Hakam Singh, Bradbury, CA (US); Ahmed Sharaby, Canyon Country, CA (US)

(73) Assignee: **PRC-DeSoto International, Inc.**, Glendale, CA (US)

(\*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 82 days.

This patent is subject to a terminal disclaimer.

(21) Appl. No.: **09/756,573**

(22) Filed: **Jan. 8, 2001**

#### Related U.S. Application Data

(63) Continuation-in-part of application No. 08/928,972, filed on Sep. 12, 1997, now Pat. No. 6,172,179, which is a continuation-in-part of application No. 08/802,130, filed on Feb. 19, 1997, now Pat. No. 5,912,319, application No. 09/756,573, which is a continuation-in-part of application No. 09/318,500, filed on May 25, 1999, now Pat. No. 6,232,401, which is a division of application No. 08/802,130, filed on Feb. 19, 1997, now Pat. No. 5,912,319, application No. 09/756,573  
(60) Provisional application No. 60/215,548, filed on Jun. 30, 2000, and provisional application No. 60/182,396, filed on Feb. 14, 2000.

(51) Int. Cl.<sup>7</sup> ..... **C08L 41/00; C08G 75/00**

(52) U.S. Cl. .... **525/212; 525/328.5; 525/328.9; 528/376; 528/373**

(58) Field of Search ..... **525/212, 328.5, 525/328.9; 528/376, 373**

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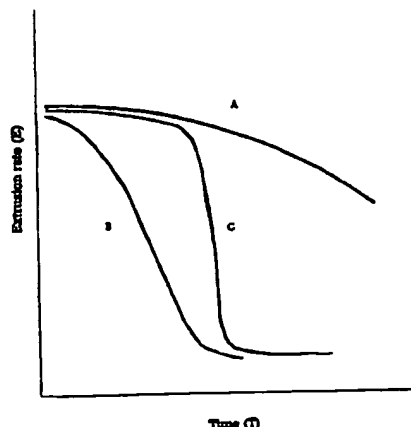
Primary Examiner—Duc Truong

(74) Attorney, Agent, or Firm—Finnegan, Henderson, Farabow, Garrett & Dunner, L.L.P.

#### (57) ABSTRACT

Sealant and potting formulations are provided which are prepared from components including ungelled mercapto-terminated polymer(s) prepared by reacting reactants comprising polyvinyl ether monomer(s) and polythiol material (s); curing agent(s) reactive with a mercapto group of the mercapto-terminated polymer; and additive(s) selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

21 Claims, 2 Drawing Sheets



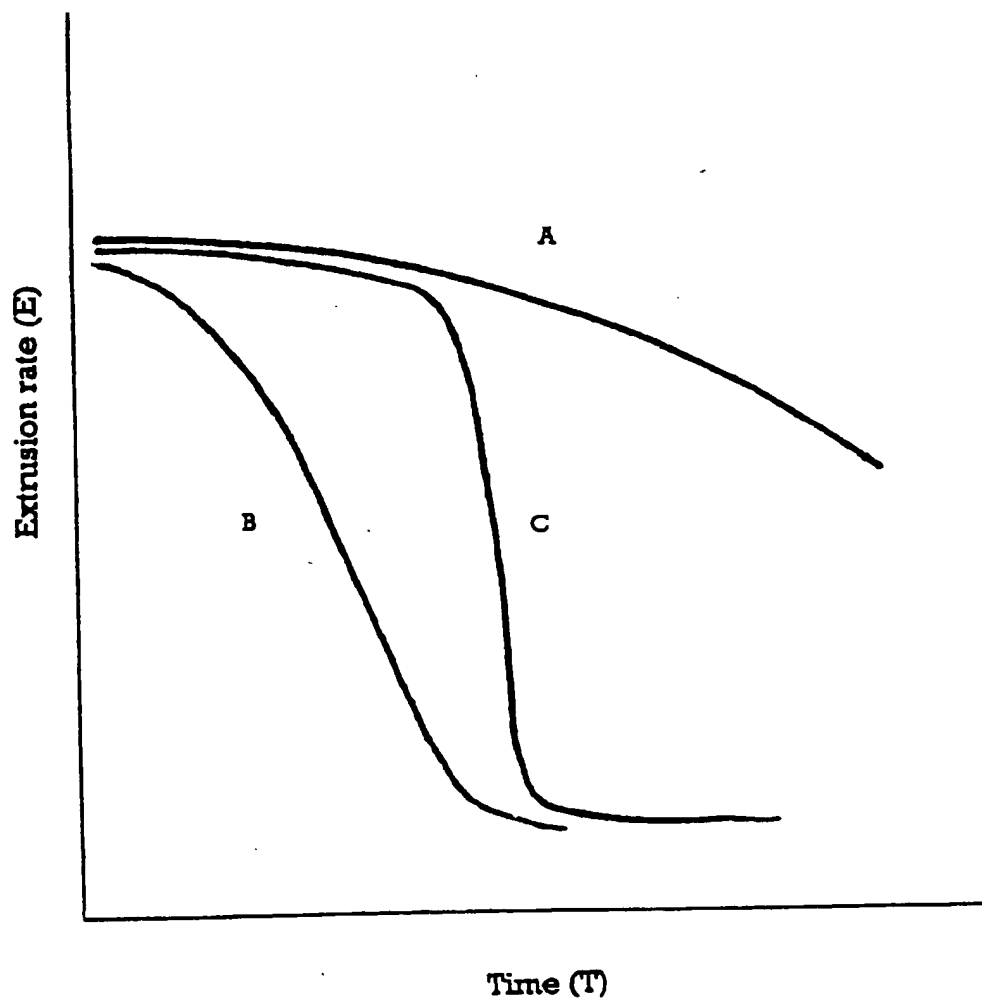
**ZOOK EXHIBIT 1014**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

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Fig. 1



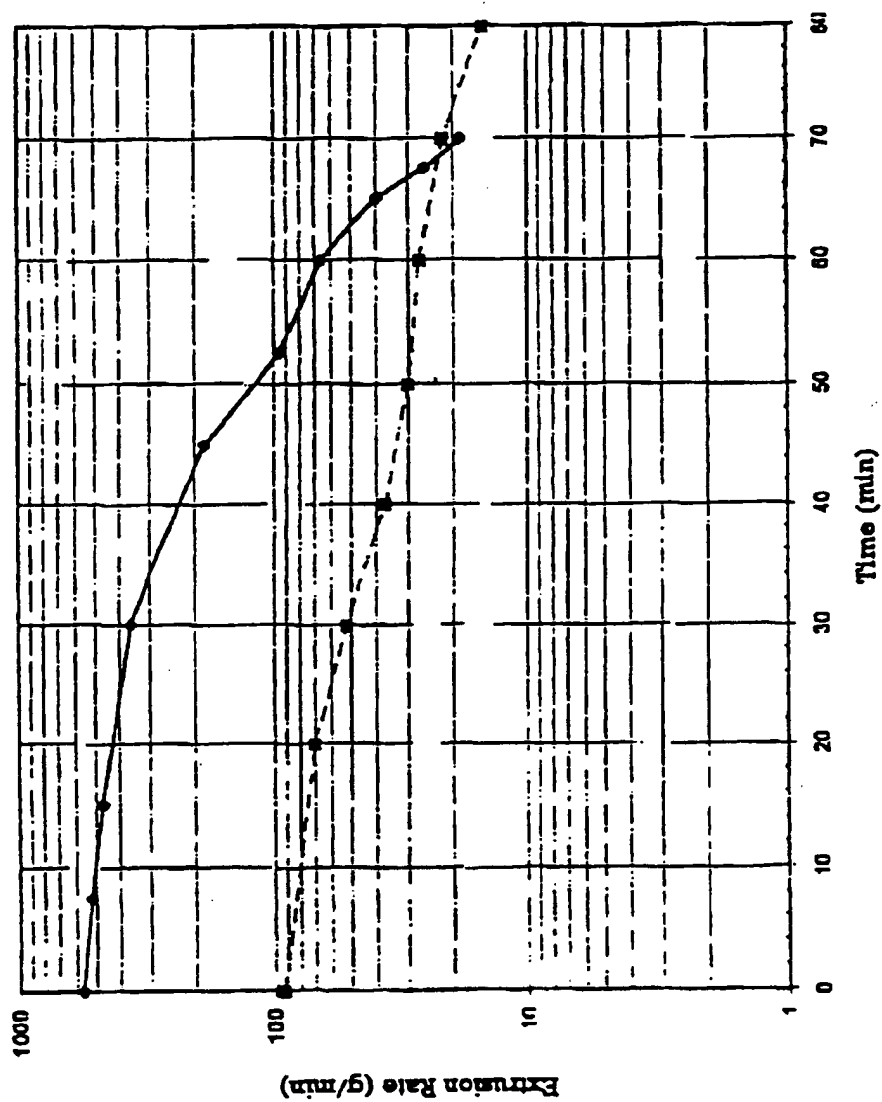


Fig. 2

# SEALANTS AND POTTING FORMULATIONS INCLUDING MERCAPTO-TERMINATED POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER

## CROSS-REFERENCE TO RELATED APPLICATIONS

This application is a continuation-in-part of U.S. patent application Ser. No. 08/928,972 filed Sep. 12, 1997 now U.S. Pat. No. 6,172,179, which is a continuation-in-part of Ser. No. 08/802,130, filed Feb. 19, 1997 now U.S. Pat. No. 5,912,319. Also, this application is a continuation-in-part of U.S. patent application Ser. No. 09/318,500 filed May 25, 1999 now U.S. Pat. No. 6,232,401 which is a division of Ser. No. 08/802,130, filed Feb. 19, 1997 now U.S. Pat. No. 5,912,319. This application also claims the benefit of U.S. provisional application No. 60/182,396 filed Feb. 14, 2000 and U.S. provisional application No. 60/215,548 filed Jun. 30, 2000.

## FIELD OF THE INVENTION

The present invention relates to a sealant or potting formulation prepared from a mercapto-terminated polymer produced by the reaction of polythiol(s) and polyvinyl ether monomer(s), the formulation having good low temperature flexibility and fuel resistance.

## BACKGROUND OF THE INVENTION

Commercially available polymeric materials which have sufficient sulfur content to exhibit desirable sealing and fuel resistance properties for aerospace sealants and electrical potting compounds are the polysulfide polyformal polymers described, e.g., in U.S. Pat. No. 2,466,963, and the alkyl side chain containing polythioether polyether polymers described, e.g., in U.S. Pat. No. 4,366,307 to Singh et al. Materials useful in this context also have the desirable properties of low temperature flexibility characterized by a low glass transition temperature ( $T_g$ ) and liquidity at room temperature.

An additional desirable combination of properties for aerospace sealants which is much more difficult to obtain is the combination of long application time (i.e., the time during which the sealant remains usable) and short curing time (the time required to reach a predetermined strength). Singh et al., U.S. Pat. No. 4,366,307, disclose such materials. Singh et al. teach the acid-catalyzed condensation of hydroxyl-functional thioethers. The hydroxyl groups are in the  $\beta$ -position with respect to a sulfur atom for increased condensation reactivity. The Singh et al. patent also teaches the use of such hydroxyl-functional thioethers with pendant methyl groups to afford polymers having good flexibility and liquidity. However, the disclosed condensation reaction has a maximum yield of about 75% of the desired condensation product. Furthermore, the acid-catalyzed reaction of  $\beta$ -hydroxysulfide monomers yields significant quantities of an aqueous solution of thermally stable and highly malodorous cyclic byproducts, such as 1-thia-4-oxa-cyclohexane which limits the suitable application of the disclosed polymers.

Another desirable feature in polymers suitable for use in aerospace sealants is high temperature resistance. While incorporating sulfur to carbon bonds into a polymer generally enhances high temperature performance, the polysulfide polyformal polymers disclosed in U.S. Pat. No. 2,466,963

have multiple —S—S— linkages in the polymer backbones which result in compromised thermal resistance. In the polymers of Singh et al., U.S. Pat. No. 4,366,307, enhanced thermal stability is achieved through replacement of polysulfide linkages with polythioether (—S—) linkages. However, the thermal resistance of these polythioethers is limited as a result of residual acid condensation catalyst.

Morris et al., U.S. Pat. No. 4,609,762, describes reacting dithiols with secondary or tertiary alcohols to afford liquid polythioethers having no oxygen in the polymeric backbone. Cured polymeric materials formed from these polymers have the disadvantage, however, of reduced fuel resistance due to the large number of pendant methyl groups that are present. In addition, the disclosed process generates undesirable aqueous acidic waste.

Cameron, U.S. Pat. No. 5,225,472, discloses production of polythioether polymers by the acid-catalyzed condensation of dithiols with active carbonyl compounds such as HCOOH. Again, this process generates undesirable aqueous acidic waste.

The addition polymerization of aromatic or aliphatic dithiols with diene monomers has been described in the literature. See, e.g., Klemm, E. et al., *J. Macromol. Sci.-Chem.*, A28(9), pp. 875-883 (1991); Nuyken, O. et al., *Makromol. Chem., Rapid Commun.* 11, 365-373 (1990). However, neither Klemm et al. nor Nuyken suggest selection of particular starting materials to form a polymer that is liquid at room temperature and, upon curing, has excellent low-temperature flexibility (low  $T_g$ ) and high resistance to fuels, i.e., hydrocarbon fluids. Nor do Klemm et al. suggest production of a polymer that also is curable at room or lower temperatures. Moreover, the reactions disclosed by Klemm et al. also generate undesirable cyclic byproducts.

There exists a need in the art for sealant, coating and electrical potting formulations or compositions that can provide good pot life as well as good performance properties, such as fuel resistance, flexural strength, thermal resistance and longevity in use.

## SUMMARY OF THE INVENTION

The present invention provides a sealant or potting formulation prepared from components comprising (a) at least one ungelled mercapto-terminated polymer prepared by reacting reactants comprising at least one polyvinyl ether monomer and at least one polythiol material; (b) at least one curing agent reactive with a mercapto group of the mercapto-terminated polymer; and (c) at least one additive selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

Another aspect of the present invention is a sealant or potting formulation prepared from components comprising: (a) at least one ungelled mercapto-terminated polymer prepared from reactants comprising diethylene glycol divinyl ether and dimercapto dioxaoctane; (b) at least one curing agent reactive with a mercapto group of the mercapto-terminated polymer; and (c) at least one additive selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

The above sealant formulations are useful in a variety of applications, such as for example aerospace applications or as electrical potting compounds.

Other than in the operating examples, or where otherwise indicated, all numbers expressing quantities of ingredients, reaction conditions, and so forth used in the specification and claims are to be understood as being qualified in all instances by the term "about". Also, as used herein, the term

"polymer" is meant to refer to oligomers, homopolymers and copolymers.

### BRIEF DESCRIPTION OF THE DRAWINGS

The invention may be more readily understood by referring to the accompanying drawings in which:

FIG. 1 depicts linear graphs of extrusion rate (E) versus time (T) for sealant compositions of the invention in comparison to extrusion rate curves for known types of sealant composition, and

FIG. 2 is a semi-log graph of the extrusion rate curve of a polythioether sealant composition of the invention (◆) and a prior art polysulfide sealant composition (■).

### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The sealant and potting formulations of the present invention comprise one or more ungelled mercapto-terminated polymers or polythioethers. It has surprisingly been discovered that polythioethers prepared from the combination of polythiol(s) with polyvinyl ether monomer(s) according to the present invention result in ungelled mercapto-terminated polymers that are liquid at room temperature and pressure and that have desirable physical and rheological properties, and that furthermore are substantially free of malodorous cyclic by-products. The inventive materials also are substantially free of deleterious catalyst residues, and have superior thermal resistance properties.

The mercapto-terminated polymers useful in the sealant and potting formulations of the present invention are preferably liquid at room temperature and pressure and cured sealants including such polymers have excellent low temperature flexibility and fuel resistance. As used herein, the term "room temperature and pressure" denotes conditions of approximately 77° F. (25° C.) and 1 atmosphere (760 mm Hg) pressure.

The mercapto-terminated polymer is ungelled or substantially free of crosslinking. By "ungelld" is meant that the mercapto-terminated polymer is substantially free of crosslinking and has an intrinsic viscosity when dissolved in a suitable solvent, as determined, for example, in accordance with ASTM-D1795 or ASTM-D4243. The intrinsic viscosity of the mercapto-terminated polymer is an indication of its finite molecular weight. A gelled reaction product, on the other hand, since it is of essentially infinitely high molecular weight, will have an intrinsic viscosity too high to measure.

Preferably, the mercapto-terminated polymer has a glass transition temperature ( $T_g$ ) that is not higher than -50° C., more preferably not higher than -55° C. and most preferably not higher than -60° C. Generally, it is preferred that the glass transition temperature of the mercapto-terminated polymer ranges from -85° C. to -50° C., and more preferably -70° C. to -50° C., as determined by differential scanning calorimetry (DSC).

Low temperature flexibility can be determined by known methods, for example, by the methods described in AMS (Aerospace Material Specification) 3267 §4.5.4.7, MIL-S (Military Specification) -8802E §3.3.12 and MIL-S-29574, and by methods similar to those described in ASTM (American Society for Testing and Materials) D522-88, which are incorporated herein by reference. Cured formulations having good low temperature flexibility are desirable in aerospace applications because the formulations are subjected to wide variations in environmental conditions, such as temperature and pressure, and physical conditions such as joint contraction and expansion and vibration.

An advantage of the formulations of the present invention is that they exhibit very desirable fuel resistance characteristics when cured, due at least in part to the use of the mercapto-terminated polymers discussed herein. The fuel resistance of a cured sealant can be determined by percent volume swell after prolonged exposure of the cured sealant to a hydrocarbon fuel, which can be quantitatively determined using methods similar to those described in ASTM D792 or AMS 3269, which are incorporated herein by reference. For fuel resistance testing, the cured sealant can be prepared from 100 parts by weight of mercapto-terminated polymer, 50 parts by weight of precipitated calcium carbonate and an epoxy curing agent in a 1:1 equivalent ratio of mercapto groups to epoxy groups. The epoxy curing agent is prepared from a 60:40 weight ratio of EPON 828 bisphenol A diglycidyl ether (available from Shell Chemical) to DEN 431 bisphenol A novolac resin (available from Dow Chemical).

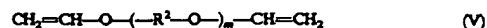
In a preferred embodiment, the cured sealants of the present invention have a percent volume swell not greater than 40%, and preferably not greater than 25% after immersion for one week at 140° F. (60° C.) and ambient pressure in jet reference fluid (JRF) type 1. More preferably, the percent volume swell of the cured polymers is not greater than 20%, and more preferably ranges from zero to 20%. Jet reference fluid JRF type 1, as employed herein for determination of fuel resistance, has the following composition (see AMS 2629, issued Jul. 1, 1989), §3.1.1 et seq., available from SAE (Society of Automotive Engineers, Warrendale, Pa.) (that is incorporated herein by reference):

Toluene	28 ± 1% by volume
Cyclohexane (technical)	34 ± 1% by volume
Isocetane	38 ± 1% by volume
Tertiary dibutyl disulfide (doctor sweet)	1 ± 0.005% by volume
Tertiary butyl mercaptan	0.015% ± 0.0015 by weight of the other four components

Preferably, the ungelled mercapto-terminated polymers have a number average molecular weight ranging from about 500 to about 20,000 grams per mole, more preferably from about 1,000 to about 10,000, and most preferably from about 2,000 to about 5,000, the molecular weight being determined by gel-permeation chromatography using a polystyrene standard.

Liquid mercapto-terminated polymers within the scope of the present invention can be difunctional, that is, linear polymers having two end groups, or polyfunctional, that is, branched polymers having three or more end groups.

The mercapto-terminated polymers are prepared by reacting reactants comprising one or more polyvinyl ether monomers and one or more polythiol materials. Useful polyvinyl ether monomers include divinyl ethers having the formula (V):



where  $\text{R}^2$  is  $\text{C}_{2-6}$  n-alkylene,  $\text{C}_{2-6}$  branched alkylene,  $\text{C}_{6-8}$  cycloalkylene or  $\text{C}_{6-10}$  alkylcycloalkylene group or  $-\{(\text{CH}_2)_p-\text{O}-\}_q-(\text{CH}_2)_r-$  and m is a rational number ranging from 0 to 10, p is an independently selected integer ranging from 2 to 6, q is an independently selected integer ranging from 1 to 5 and r is an independently selected integer ranging from 2 to 10.

The materials of formula V are divinyl ethers. Such divinyl ether monomers as described herein can provide

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polymers having superior fuel resistance and low temperature performance as compared to prior art polymers prepared from alkenyl ether and conjugated dienes such as 1,3-butadiene copolymerized with a dithiol such as DMDS. Divinyl ether ( $m=0$ ) is operative herein. Preferred divinyl ethers include those compounds having at least one oxyalkylene group, more preferably from 1 to 4 oxyalkylene groups such as those compounds in which  $m$  is an integer from 1 to 4. More preferably,  $m$  is an integer from 2 to 4. It is also possible to employ commercially available divinyl ether mixtures in producing mercapto-terminated polymers according to the invention. Such mixtures are characterized by a non-integral average value for the number of alkoxy units per molecule. Thus,  $m$  in formula V can also take on rational number values between 0 and 10.0; preferably between 1.0 and 10.0; very preferably between 1.0 and 4.0, particularly between 2.0 and 4.0.

Suitable polyvinyl ether monomers include divinyl ether monomers, such as divinyl ether, ethylene glycol divinyl ether (EG-DVE) ( $R^2$ =ethylene,  $m=1$ ), butanediol divinyl ether (BD-DVE) ( $R^2$ =butylene,  $m=1$ ), hexanediol divinyl ether (HD-DVE) ( $R^2$ =hexylene,  $m=1$ ), diethylene glycol divinyl ether (DEG-DVE) ( $R^2$ =ethylene,  $m=2$ ) (preferred), triethylene glycol divinyl ether ( $R^2$ =ethylene,  $m=3$ ), tetraethylene glycol divinyl ether ( $R^2$ =ethylene,  $m=4$ ), cyclohexanedimethanol divinyl ether, polytetrahydrofuryl divinyl ether; trivinyl ether monomers such as trimethylolpropane trivinyl ether, tetrafunctional monomers such as pentaerythritol tetrafunctional ether and mixtures thereof. The polyvinyl ether material can have one or more pendant groups selected from alkyl groups, hydroxyl groups, alkoxy groups and amine groups.

Useful divinyl ethers in which  $R^2$  is  $C_{2-6}$  branched alkylene can be prepared by reacting a polyhydroxy compound with acetylene. Exemplary compounds of this type include compounds in which  $R^2$  is an alkyl-substituted methylene group such as  $-\text{CH}(\text{CH}_3)-$  (for example "PLURIOL®" blends such as PLURIOL®E-200 divinyl ether (BASF Corp. of Parsippany, N.J.), for which  $R^2$ =ethylene and  $m=3.8$ ) or an alkyl-substituted ethylene (for example  $-\text{CH}_2\text{CH}(\text{CH}_3)-$  such as "DPE" polymeric blends including DPE-2 and DPE-3 (International Specialty Products of Wayne, N.J.)).

Other useful divinyl ethers include fluorinated compounds or compounds in which  $R^2$  is polytetrahydrofuryl (poly-THF) or polyoxyalkylene, preferably having an average of about 3 monomer units.

Two or more polyvinyl ether monomers of the formula V can be used in the foregoing method. Thus in preferred embodiments of the invention, two polythiols of formula IV (discussed below) and one polyvinyl ether monomer of formula V, one polythiol of formula IV and two polyvinyl ether monomers of formula V, two polythiols of formula IV and two polyvinyl ether monomers of formula V, and more than two compounds of one or both formulas, can be used to produce a variety of polymers according to the invention, and all such combinations of compounds are contemplated as being within the scope of the invention.

Generally, the polyvinyl ether monomer comprises 20 to less than 50 mole percent of the reactants used to prepare the mercapto-terminated polymer, and preferably 30 to less than 50 mole percent.

Suitable polythiol materials for preparing the mercapto-terminated polymer include compounds, monomers or polymers having at least two thiol groups. Useful polythiols include dithiols having the formula (IV):



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where  $R^1$  can be a  $C_{2-6}$   $n$ -alkylene group;  $C_{3-6}$  branched alkylene group, having one or more pendant groups which can be, for example, hydroxyl groups, alkyl groups such as methyl or ethyl groups; alkoxy groups,  $C_{6-8}$  cycloalkylene;  $C_{4-10}$  alkylcycloalkylene group;  $-\{(-\text{CH}_2)_p-\text{X}\}_q-$  ( $-\text{CH}_2$ ),—; or  $-\{(-\text{CH}_2)_p-\text{X}\}_q-$  ( $-\text{CH}_2$ ),— in which at least one  $-\text{CH}_2-$  unit is substituted with a methyl group and in which  $p$  is an independently selected integer ranging from 2 to 6,  $q$  is an independently selected integer ranging from 1 to 5 and  $r$  is an independently selected integer ranging from 2 to 10.

Further preferred dithiols include one or more heteroatom substituents in the carbon backbone, that is, dithiols in which  $X$  includes a heteroatom such as O, S or another bivalent heteroatom radical; a secondary or tertiary amine group, i.e.,  $-\text{NR}^6-$ , where  $R^6$  is hydrogen or methyl; or another substituted trivalent heteroatom. In a preferred embodiment,  $X$  is O or S, and thus  $R^1$  is  $-\{(-\text{CH}_2)_p-\text{O}-\}_q-$  ( $-\text{CH}_2$ ),— or  $-\{(-\text{CH}_2)_p-\text{S}-\}_q-$  ( $-\text{CH}_2$ ),—. Preferably,  $p$  and  $r$  are equal, and most preferably both have the value of 2.

Useful polythiols include but are not limited to dithiols such as 1,2-ethanedithiol, 1,2-propanedithiol, 1,3-propanedithiol, 1,3-butanedithiol, 1,4-butanedithiol, 2,3-butanedithiol, 1,3-pentanedithiol, 1,5-pentanedithiol, 1,6-hexanedithiol, 1,3-dimercapto-3-methylbutane, dipentenedimercaptan, ethylcyclohexyldithiol (ECHDT), dimercaptodiethylsulfide, methyl-substituted dimercaptodiethylsulfide, dimethyl-substituted dimercaptodiethylsulfide, dimercaptodioxoctane, 1,5-dimercapto-3-oxapentane and mixtures thereof. The polythiol material can have one or more pendant groups selected from lower alkyl groups, lower alkoxy groups and hydroxyl groups. Suitable alkyl pendant groups include  $C_1$ – $C_6$  linear alkyl,  $C_3$ – $C_6$  branched alkyl, cyclopentyl, and cyclohexyl.

Preferred dithiols include dimercaptodiethylsulfide (DMDS) ( $p=2$ ,  $r=2$ ,  $q=1$ ,  $X=\text{S}$ ); dimercaptodioxoctane (DMDO) ( $p=2$ ,  $q=2$ ,  $r=2$ ,  $X=\text{O}$ ); and 1,5-dimercapto-3-oxapentane ( $p=2$ ,  $r=2$ ,  $q=1$ ,  $X=\text{O}$ ). It is also possible to use dithiols that include both heteroatom substituents in the carbon backbone and pendant alkyl groups, such as methyl groups. Such compounds include methyl-substituted DMDS, such as  $\text{HS}-\text{CH}_2\text{CH}(\text{CH}_3)-\text{S}-\text{CH}_2\text{CH}_2-\text{SH}$ ,  $\text{HS}-\text{CH}(\text{CH}_3)\text{CH}_2-\text{S}-\text{CH}_2\text{CH}_2-\text{SH}$  and dimethyl substituted DMDS such as  $\text{HS}-\text{CH}_2\text{CH}(\text{CH}_3)-\text{S}-\text{CH}(\text{CH}_3)\text{CH}_2-\text{SH}$  and  $\text{HS}-\text{CH}(\text{CH}_3)\text{CH}_2-\text{S}-\text{CH}_2\text{CH}(\text{CH}_3)-\text{SH}$ .

Two or more different polythiols can be used if desired to prepare useful polythioethers.

Preferably, the polythiol material has a number average molecular weight ranging from 90 to 1000 grams per mole, and more preferably 90 to 500 grams per mole.

Relative amounts of dithiol and divinyl ether materials used to prepare the polymers are chosen to yield terminal mercapto groups ( $-\text{SH}$ ). These mercapto-terminated polymers can include thiol terminal groups that are not further reacted ("uncapped"), or include one or more thiol groups that are further reacted with other materials to provide reactive or non-reactive terminal or pendant groups ("capped"). Capping the polymers of the present invention enables introduction of additional terminal functionalities, for example, hydroxyl or amine groups, to the polymers, or in the alternative, introduction of end groups that resist further reaction, such as terminal alkyl groups. Preferably, the stoichiometric ratio of polythiol to divinyl ether materials is less than one equivalent of polyvinyl ether to one equivalent of polythiol, resulting in mercapto-terminated

polymers. More preferably, the stoichiometric ratio is selected to fully terminate the polymer with mercapto groups.

Hydroxyl- or amino-functional terminal polymers can be produced, for example, by reacting a vinyl terminated material with mercaptoalcohols such as 3-mercaptopropanol or mercaptoamines such as 4-mercaptobutylamine, respectively, or by reacting a mercaptan terminated material with a vinyl terminated material having hydroxyl functionality such as butane diol monovinyl ether or amine functionality such as aminopropyl vinyl ether.

Preferably, the mercapto-terminated polymer comprises 30 to 90 weight percent of the sealant formulation on a basis of total weight of the sealant formulation, and more preferably 30 to 60 weight percent.

The reactants from which the mercapto-terminated polymers are prepared can further comprise one or more free radical catalysts. Preferred free radical catalysts include azo compounds, for example azobis-nitrile compounds such as azo(bis)isobutyronitrile (AIBN); organic peroxides such as benzoyl peroxide and t-butyl peroxide; inorganic peroxides and similar free-radical generators. The reaction can also be effected by irradiation with ultraviolet light either with or without a cationic photoinitiating moiety. Ionic catalysis methods, using either inorganic or organic bases, e.g., triethylamine, also yield materials useful in the context of this invention.

Mercapto-terminated polymers within the scope of the present invention can be prepared by a number of methods. According to a first preferred method, (n+1) moles of a material having the formula IV:



or a mixture of at least two different compounds having the formula IV, are reacted with n moles of a material having the formula V:



or a mixture of at least two different compounds having the formula V, in the presence of a catalyst. This method provides an uncapped, mercapto-terminated difunctional polymer.

Although, as indicated above, compounds of the formulas IV and V which have pendant alkyl groups, for example pendant methyl groups, are useful according to the invention, it has surprisingly been discovered that compounds of the formulas IV and V which are free of pendant methyl or other alkyl groups also afford mercapto-terminated polymers that are ungelled at room temperature and pressure.

Capped analogs to the foregoing mercapto-terminated polymers can be prepared by reacting a material having the formula IV or a mixture of at least two different compounds having the formula IV and a material having the formula V or a mixture of at least two different compounds having the formula V in a stoichiometric ratio of less than one equivalent of dithiol per vinyl equivalent of formula V, with about 0.05 to about 2 moles of a material having the formula VI



or a mixture of two different materials having the formula VI, in the presence of an appropriate catalyst.

Materials of the formula VI are alkyl  $\omega$ -alkenyl ethers having a terminal ethylenically unsaturated group which react with terminal thiol groups to cap the polythioether polymer.

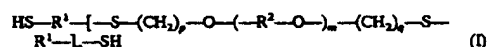
In formula VI, s is an integer from 0 to 10, preferably 0 to 6, more preferably 0 to 4 and  $\text{R}^5$  is an unsubstituted or substituted alkyl group, preferably a  $\text{C}_{1-6}$  n-alkyl group which can be substituted with at least one  $-\text{OH}$  or  $-\text{NHR}^7$  group, with  $\text{R}^7$  denoting H or  $\text{C}_{1-6}$  alkyl. Exemplary useful  $\text{R}^5$  groups include alkyl groups, such as ethyl, propyl and butyl, hydroxyl-substituted groups such as 4-hydroxybutyl; amine substituted groups such as 3-aminopropyl; etc.

Specific preferred materials of the formula VI are monovinyl ethers (s=0), including amino- and hydroxyalkyl vinyl ethers, such as 3-aminopropyl vinyl ether and 4-hydroxybutyl vinyl ether (butanediol monovinyl ether), as well as unsubstituted alkyl vinyl ethers such as ethyl vinyl ether. Additional preferred materials of the formula VI include allyl ethers (s=1), such as 4-aminobutyl allyl ether, 3-hydroxypropyl allyl ether, etc. Although materials in which s is greater than 6 can be used, the resulting polymers may have less fuel resistance than those in which s is 6 or less.

Use of equivalent amounts of materials of the formula VI relative to thiol groups present in formula IV provides fully capped mercapto polymers, while use of lesser amounts results in partially capped polymers.

Preferably, an equivalent of polyvinyl ether is reacted with dithiol or a mixture of polythiols.

A preferred linear structured mercapto-terminated polymer useful in the sealant and potting formulations of the present invention has the structure of formula (I):



wherein

$\text{R}^1$  denotes a  $\text{C}_{2-10}$  n-alkylene,  $\text{C}_{2-6}$  branched alkylene,  $\text{C}_{6-8}$  cycloalkylene or  $\text{C}_{6-10}$  alkylcycloalkylene group, heterocyclic,  $-\{(\text{CH}_2)_p-\text{X}\}_q-(\text{CH}_2)_r-$ ; or  $-\{(\text{CH}_2)_p-\text{X}\}_q-(\text{CH}_2)_r-$  in which at least one  $-\text{CH}_2-$  unit is substituted with a methyl group;

$\text{R}^2$  denotes a  $\text{C}_{2-10}$  n-alkylene,  $\text{C}_{2-6}$  branched alkylene,  $\text{C}_{6-8}$  cycloalkylene or  $\text{C}_{6-14}$  alkylcycloalkylene group, heterocyclic,  $-\{(\text{CH}_2)_p-\text{X}\}_q-(\text{CH}_2)_r-$ ;

X denotes one selected from the group consisting of O, S and  $-\text{NR}^6-$ ;

$\text{R}^6$  denotes H or methyl;

m is an independently selected rational number from 1 to 50; and

n is an independently selected integer from 1 to 60;

p is an independently selected integer ranging from 2 to 6;

q is an independently selected integer ranging from 1 to 5; and

r is an independently selected integer from 2 to 10.

In a more preferred embodiment of the foregoing polymer,  $\text{R}^1$  is  $\text{C}_2-\text{C}_6$  alkyl and  $\text{R}^2$  is  $\text{C}_2-\text{C}_6$  alkyl.

In a preferred embodiment, the polythioether has the formula (II):



wherein

A denotes a structure having the formula I,

y is 0 or 1,

$\text{R}^3$  denotes a single bond when y=0 and  $-\text{S}-(\text{CH}_2)_2-[\text{O}-\text{R}^2]_m-\text{O}-$  when y=1,

$\text{R}^4$  denotes  $-\text{SH}$  or  $-\text{S}-(\text{CH}_2)_2-\text{O}-\text{R}^5$  when y=0 and  $-\text{CH}=\text{CH}_2$  or  $-(\text{CH}_2)_2-\text{S}-\text{R}^5$  when y=1,

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s is an integer from 0 to 10,

R<sup>5</sup> denotes C<sub>1-6</sub> alkyl group which is unsubstituted or substituted with at least one —OH or —NHR<sup>7</sup> group, and

R<sup>7</sup> denotes H or a C<sub>1-6</sub> n-alkyl group.

Thus, polythioethers of the formula II are linear, difunctional polymers which can be uncapped or capped. When y=0, the polymer includes terminal thiol groups or capped derivatives thereof. In an alternative embodiment, when y=1 (not preferred), the polymer includes terminal vinyl groups or capped derivatives thereof.

According to a preferred embodiment, the inventive polythioether is a difunctional thiol-terminated (uncapped) polythioether. That is, in formula II, y=0 and R<sup>4</sup> is —SH. Thus, the polythioether has the following structure: HS—R<sup>1</sup>—[—S—(CH<sub>2</sub>)<sub>2</sub>—O—[—R<sup>2</sup>—O—]<sub>m</sub>—(CH<sub>2</sub>)<sub>2</sub>—S—R<sup>1</sup>—]<sub>n</sub>—SH

In a preferred embodiment, R<sup>1</sup>=[—(CH<sub>2</sub>)<sub>p</sub>—X]<sub>q</sub>—(—CH<sub>2</sub>)<sub>r</sub>—, where p=2, X=O, q=2 and r=2, R<sup>2</sup> is ethylene group, m=2 and n is about 9.

The foregoing polymers are produced, for example, by reacting a divinyl ether or mixture thereof with an excess of a dithiol or mixture thereof, as discussed in detail below.

In an alternative embodiment of the foregoing polythioether, when m=1 and R<sup>2</sup>=n-butylene in formula II, R<sup>1</sup> is not ethylene or n-propylene. Also preferably, when m=1, p=2, q=2, r=2 and R<sup>2</sup>=ethylene, X is not O.

Although not preferred, polythioethers according to the invention can also include difunctional vinyl-terminated polythioethers. That is, in formula II, y=1 and R<sup>4</sup> is —CH=CH<sub>2</sub>. These polymers are produced, for example, by reacting a dithiol or mixture thereof with an excess of a divinyl ether or mixture thereof, as discussed in detail below. Analogous capped polythioethers include terminal —(CH<sub>2</sub>)<sub>2</sub>—S—R<sup>3</sup>.

Preferably, the mercapto-terminated polymers are essentially free of sulfone, ester or disulfide linkages, and more preferably free of such linkages. The absence of these linkages can provide good fuel and temperature resistance and good hydrolytic stability. As used herein, "essentially free of sulfone, ester or disulfide linkages" means that less than 2 mole percent of the linkages in the mercapto-terminated polymer are sulfone, ester or disulfide linkages. Disulfide linkages are particularly susceptible to thermal degradation, sulfone linkages are particularly susceptible to hydrolytic degradation.

Mercapto-terminated polymers useful in the formulations of the present invention have a mercapto functionality of at least 2. Polyfunctional analogs of the foregoing difunctional mercapto-terminated polymers can be prepared by reacting one or more compounds of formula IV and one or more compounds of formula V, in appropriate amounts, with one or more polyfunctionalizing agents.

The term "polyfunctionalizing agent" as employed herein denotes a compound having more than two moieties that are reactive with —SH and/or —CH=CH<sub>2</sub> groups. The polyfunctionalizing agent preferably includes from 3 to 6 such moieties, and thus is denoted a "z-valent" polyfunctionalizing agent, where z is the number (preferably from 3 to 6) of such moieties included in the agent, and hence the number of separate branches which the polyfunctional mercapto-terminated polymer comprises.

The polyfunctionalizing agent can be represented by the formula

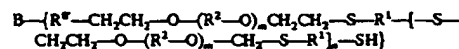


where R<sup>6</sup> denotes a moiety or several moieties that are reactive with —SH or —CH=CH<sub>2</sub> groups, and B is the

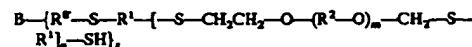
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z-valent residue of the polyfunctionalizing agent, i.e., the portion of the agent other than the reactive moieties R<sup>6</sup>.

Polyfunctional mercapto-terminated polymers according to the present invention thus preferably have the formula:



or



wherein

B denotes a z-valent residue of a polyfunctionalizing agent,

R<sup>1</sup>, R<sup>2</sup>, n and m denote structures and values discussed above with reference to Formula I,

R<sup>6</sup> denotes a moiety which is reactive with a terminal vinyl group or mercapto group, and

z is an integer from 3 to 6.

Polyfunctional polythioethers according to the present invention can preferably have the formula (III):



wherein

A denotes a structure having the formula I,

Y is 0 or 1,

R<sup>3</sup> denotes a single bond when y=0 and —S—(CH<sub>2</sub>)<sub>2</sub>—[—O—R<sup>2</sup>—]<sub>m</sub>—O— when y=1,

R<sup>4</sup> denotes —SH or —S—(—CH<sub>2</sub>)<sub>2</sub>—O—R<sup>5</sup> when y=0 and —CH=CH<sub>2</sub> or —(CH<sub>2</sub>)<sub>2</sub>—S—R<sup>5</sup> when y=1,

R<sup>5</sup> denotes C<sub>1-6</sub> alkyl that is unsubstituted or substituted with at least one —OH or —NHR<sup>7</sup> group,

R<sup>7</sup> denotes H or a C<sub>1-6</sub> n-alkyl group,

s is an integer from 0 to 10,

z is an integer from 3 to 6, and

B denotes a z-valent residue of a polyfunctionalizing agent.

As with the preceding difunctional embodiments, the foregoing polyfunctional polythioethers of the present invention optionally include terminal —SH or —CH=CH<sub>2</sub> groups, or are capped and thus include terminal —S—(—CH<sub>2</sub>)<sub>2</sub>—O—R<sup>5</sup> or —(CH<sub>2</sub>)<sub>2</sub>—S—R<sup>5</sup> groups. Partially capped polyfunctional polymers, i.e., polymers in which some but not all of the branches are capped, are also within the scope of the present invention.

Specific polyfunctionalizing agents include trifunctionalizing agents, that is, compounds with z=3. Preferred trifunctionalizing agents include triallylcyanurate (TAC), which is reactive with dithiol, and 1,2,3-propanetrithiol, which is reactive with polyvinyl ether. Agents having mixed functionality, i.e., agents that include moieties which are typically separate moieties that react with both thiol and vinyl groups, can also be employed.

Other useful polyfunctionalizing agents include trimethylolpropane trivinyl ether, and the polythiols described in U.S. Pat. Nos. 4,366,307; 4,609,762 and 5,225,472, the disclosures of each of which are incorporated in their entirety herein by reference. Mixtures of polyfunctionalizing agents can also be used.

Polyfunctionalizing agents having more than three reactive moieties (i.e., z>3) afford "star" polymers and hyperbranched polymers. For example, two moles of TAC can be

reacted with one mole of a dithiol to afford a material having an average functionality of 4. This material can then be reacted with a diene and a dithiol to yield a polymer, which can in turn be mixed with a trifunctionalizing agent to afford a polymer blend having an average functionality between 3 and 4.

Inventive polymers as described above have a wide range of average functionality. For example, trifunctionalizing agents afford average functionalities from about 2.05 to 3.0, preferably about 2.1 to 2.6. Wider ranges of average functionality can be achieved by using tetrafunctional or higher polyfunctionalizing agents. Functionality will also be affected by factors such as stoichiometry, as is known to those skilled in the art.

It is contemplated that other functional groups may be employed as a substitute for the thiol groups discussed herein to react with the curing agent in order to form the polyfunctional material of the present invention. These functional groups include, for example, hydroxyl functional groups and amine groups. These thiol substitutes may be employed in the reaction chemistry by one of ordinary skill in the art of sealant formation based upon the examples and methodology discussed herein.

Thus, according to one method for making polyfunctional polymers of the present invention, (n+1) moles of a compound or compounds having the formula IV, (n) moles of a compound or compounds having the formula V, and a z-valent polyfunctionalizing agent in an amount sufficient to obtain a predetermined molecular weight and functionality, are combined to form a reaction mixture. The mixture is then reacted in the presence of a suitable catalyst as described above to afford mercapto-terminated polyfunctional polymers. Capped analogs of the foregoing mercapto-terminated polyfunctional polymers are prepared by inclusion in the starting reaction mixture of about 0.05 to about (z) moles one or more appropriate capping compounds VI. Use of (z) moles affords fully capped polyfunctional polymers, while use of lesser amounts again yields partially capped polymers.

The inventive polymers preferably are prepared by combining at least one compound of formula IV and at least one compound of formula V, optionally together with one or more capping compounds VI and/or VII as appropriate, and/or a polyfunctionalizing agent, followed by addition of an appropriate catalyst, and carrying out the reaction at a temperature from about 50 to about 120° C. for a time from about 2 to about 24 hours. Preferably, the reaction is carried out at a temperature from about 70 to about 90° C. for a time from about 2 to about 6 hours.

Since the inventive reaction is an addition reaction, rather than a condensation reaction, the reaction typically proceeds substantially to completion, i.e., the inventive mercapto-terminated polymers are produced in yields of approximately 100%. No or substantially no undesirable by-products are produced. In particular, the reaction does not produce appreciable amounts of malodorous cyclic by-products such as are characteristic of several known methods for producing polythioethers. Moreover, the polymers prepared according to the invention are substantially free of residual catalyst.

Methods of making the foregoing polyfunctional inventive polymers are discussed in detail below.

Preferably, the mercapto-terminated polymer has a viscosity of less than about 500 poise at a temperature of about 25° C. and a pressure of about 760 mm Hg determined according to ASTM D-2849 §79-90 using a Brookfield viscometer.

The mercapto-terminated polymer or combination of mercapto-terminated polymers as detailed herein preferably is present in the polymerizable sealant composition in an amount from about 30 wt % to about 90 wt %, more preferably about 40 to about 80 wt %, very preferably about 45 to about 75 wt %, with the wt % being calculated based on the weight of total solids of the composition.

The sealant or potting formulations of the present invention further comprise one or more curing agents, such as polyolefins, polyacrylates, metal oxides, polyepoxides and mixtures thereof as appropriate. Curing agents useful in polymerizable sealant compositions of the invention include polyepoxides or epoxy functional resins, for example, hydantoin diepoxide, bisphenol-A epoxides, bisphenol-F epoxides, novolac type epoxides, aliphatic polyepoxides, and any of the epoxidized unsaturated and phenolic resins. Other useful curing agents include unsaturated compounds such as acrylic and methacrylic esters of commercially available polyols, unsaturated synthetic or naturally occurring resin compounds, TAC, and olefinic terminated derivatives of the compounds of the present invention. In addition, useful cures can be obtained through oxidative coupling of the thiol groups using organic and inorganic peroxides (e.g.,  $\text{MnO}_2$ ) known to those skilled in the art. Selection of the particular curing agent may affect the  $T_g$  of the cured composition. For example, curing agents that have a  $T_g$  significantly lower than the  $T_g$  of the polythioether may lower the  $T_g$  of the cured composition.

Depending on the nature of the mercapto-terminated polymer(s) used in the composition, the composition can comprise about 90% to about 150% of the stoichiometric amount of the selected curing agent(s) based upon —SH equivalents, preferably about 95 to about 125%.

Fillers useful in the polymerizable compositions of the invention for aerospace application include those commonly used in the art, such as carbon black and calcium carbonate ( $\text{CaCO}_3$ ). Potting compound fillers illustratively include high band gap materials such as zinc sulfide and inorganic barium compounds. Preferably, the compositions include about 10 to about 70 wt % of the selected filler or combination of fillers, more preferably about 10 to 50 wt % based upon the total weight of the composition.

The sealant and potting compositions of the present invention can comprise one or more adhesion promoters. Suitable adhesion promoters include phenolics such as METHYLON phenolic resin available from Occidental Chemicals, organosilanes such as epoxy, mercapto or amino functional silanes such as A-187 and A-1100 available from OSi Specialties. Preferably, an adhesion promoter is employed in an amount from 0.1 to 15 wt % based upon total weight of the formulation.

Common substrates to which the sealant compositions of the present invention are applied can include titanium, stainless steel, aluminum, anodized, primed, organic coated and chromate coated forms thereof, epoxy, urethane, graphite, fiberglass composite, KEVLAR®, acrylics and polycarbonates.

Preferably, a plasticizer is present in the sealant formulation in an amount ranging from 1 to 8 weight percent based upon total weight of the formulation. Plasticizers that are useful in polymerizable compositions of the invention include phthalate esters, chlorinated paraffins, hydrogenated terphenyls, etc.

The formulation can further comprise one or more organic solvents, such as isopropyl alcohol, in an amount ranging from 0 to 15 percent by weight on a basis of total weight of the formulation, preferably less than 15 weight percent and more preferably less than 10 weight percent.

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A typical sealant formulation is provided in Example 18.

Polymerizable sealant composition cure time is reduced considerably by using an organic amine catalyst having a pKb of 10 or above. Preferred organic amine catalysts are organic tertiary amines. Specific catalysts which are useful in the present invention are triethylene diamine, diazabicyclo (2,2,2) octane (DABCO) (preferred), diazabicycloundecene (DBU), 2,4,6-tri(dimethylamino methyl) phenol (DMP-30) and tetramethyl guanidine (TMG). The reaction time when utilizing the organic amine catalysts, and particularly the organic tertiary amine catalysts, is in general between about one hour to about 20 hours which is a considerable difference compared to using no amine catalyst.

Generally the amount of catalyst ranges from 0.05 wt % to 3 wt %, based on the total weight of the starting reactants.

The foregoing sealant or potting formulations preferably are cured at ambient temperature and pressure, however the formulations generally can be cured at a temperature ranging from about 0° C. to about 100° C.

In addition to the foregoing ingredients, polymerizable sealant compositions of the invention can optionally include one or more of the following: pigments; thixotropes; retardants; and masking agents.

Useful pigments include those conventional in the art, such as carbon black and metal oxides. Pigments preferably are present in an amount from about 0.1 to about 10 wt % based upon total weight of the formulation.

Thixotropes, for example fumed silica or carbon black, are preferably used in an amount from about 0.1 to about 5 wt % based upon total weight of the formulation.

An additional advantage of sealant formulations according to the invention is their improved curing behavior. The extent of cure of a sealant formulation as a function of time is often difficult to measure directly, but can be estimated by determining the extrusion rate of the composition as a function of time. The extrusion rate is the rate at which a mixed sealant formulation, i.e., a sealant formulation together with an accelerator system, is extruded from an applicator device. As the sealant formulation is mixed with the accelerator system, curing begins, and the extrusion rate changes with time. The extrusion rate thus is inversely related to the extent of cure. When the extent of cure is low, the viscosity of the mixed ungelled sealant formulation is low and thus the extrusion rate is high. When the reaction approaches completion, the viscosity becomes very high, and the extrusion rate thus becomes low. The extrusion rate can be measured according to AMS Method 3276 (section 4.5.10), which is incorporated herein by reference.

With reference to FIG. 1, the viscosity of some known types of sealant formulations remains low for an extended time, because the compositions are slow to cure. Such formulations have extrusion curves qualitatively similar to curve A. Other known types of sealant formulations cure very quickly, and thus their viscosity rapidly increases. Consequently, the extrusion rate rapidly decreases, as shown in curve B. Desirably, a mixed sealant formulation should have a low viscosity, and thus a high extrusion rate, for a length of time sufficient to allow even application of the sealant formulation to the area requiring sealing, but then should cure rapidly after application, i.e., their extrusion rate should quickly decrease. Sealant formulations according to the present invention are characterized by this desirable extrusion curve, as illustrated qualitatively in curve C.

Sealant formulations according to the present invention can have, depending on the particular formulation, initial extrusion rates as high as 500 g/min or higher, together with

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low extrusion rates on the order of about 5 to 10 g/min or less after curing times on the order of one hour.

As shown in FIG. 2, the initial extrusion rate of a sealant containing a polymer of the present invention (Example 1, below, cured with an epoxy curing agent as described below) is about 550 g/min, then falls rapidly to about 20 g/min after 70 minutes. In comparison, a known polysulfide polymer based sealant (cured with MnO<sub>2</sub>) has an initial extrusion rate of about 90 g/min, which slowly falls to about 20 g/min after 70 minutes.

Another preferred curable sealant formulation combines one or more plasticizers with the mercapto-terminated polymer(s), curing agent(s) and filler(s) described above. Use of a plasticizer allows the polymerizable formulation to include mercapto-terminated polymers which have higher T<sub>g</sub> than would ordinarily be useful in an aerospace sealant or potting compound, i.e., use of a plasticizer effectively reduces the T<sub>g</sub> of the formulation, and thus increases the low-temperature flexibility of the cured polymerizable formulation beyond that which would be expected on the basis of the T<sub>g</sub> of the mercapto-terminated polymers alone.

The present invention is illustrated in more detail by means of the following non-limiting examples which are presently representative of preferred embodiments. These examples are exemplary and are not intended as a limitation on the scope of the invention as detailed in the appended claims.

## EXAMPLES

In Examples 1-8, liquid polythioethers were prepared by stirring together one or more dithiols with one or more divinyl ethers and a trifunctionalizing agent. The reaction mixture was then heated and a free radical catalyst was added. All reactions proceeded substantially to completion (approximately 100% yield).

### Example 1

In a 2 L flask, 524.8 g (3.32 mol) of diethylene glycol divinyl ether (DEG-DVE) and 706.7 g (3.87 mol) of dimercaptodioxaoctane (DMDO) were mixed with 19.7 g (0.08 mol) of triallylcyanurate (TAC) and heated to 77° C. To the heated reaction mixture was added 4.6 g (0.024 mol) of an azobisnitrile free radical catalyst (VAZO® 67 [2,2-azobis (2-methylbutyronitrile), commercially available from DuPont]. The reaction proceeded substantially to completion after 2 hours to afford 1250 g (0.39 mol, yield 100%) of a liquid polythioether resin having a T<sub>g</sub> of -68° C. and a viscosity of 65 poise and a number average molecular weight of about 3165 grams per mole and a thiol functionality of 2.2. The polymer was faintly yellow and had low odor.

### Example 2

In a 1 L flask, 404.4 g (1.60 mol) of PLURIOL® E-200 divinyl ether and 355.88 g (1.94 mol) of DMDO were mixed with 12.1 g (0.049 mol) of TAC and reacted as in Example 1. The reaction proceeded substantially to completion after 5 hours to afford 772 g (0.024 mol, yield 100%) of a resin having a T<sub>g</sub> of -66° C. and a viscosity of 48 poise. The polymer was yellow and had low odor.

### Example 3

In a 100 mL flask, 33.2 g (0.21 mol) of DEG-DVE and 26.48 g (0.244 mol) of 1,2-propanedithiol were mixed with 0.75 g (0.003 mol) of TAC and heated to 71° C. To the

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heated reaction mixture was added 0.15 g (0.8 mmol) of VAZO® 67. The reaction proceeded substantially to completion after 7 hours to afford 60 g (0.03 mol, yield 100%) of a resin having a  $T_g$  of  $-61^\circ\text{C}$ . and a viscosity of 22 poise. The polymer had a noticeable PDT (propane s dithiol) odor.

## Example 4

In a 100 mL flask, 33.3 g (0.136 mol) of tripropylene glycol divinyl ether (DPE-3) and 27.0 g (0.170 mol) of dimercaptodiethylsulfide (DMDS) were mixed with 0.69 g (0.003 mol) of TAC and heated to  $77^\circ\text{C}$ . To the heated reaction mixture was added 0.15 g (0.8 mmol) of VAZO® 67. The reaction proceeded substantially to completion after 6 hours to afford 61 g (0.028 mol, yield 100%) of a polymer having a  $T_g$  of  $-63^\circ\text{C}$ . and a viscosity of 26 poise.

## Example 5

In a 250 mL flask, 113.01 g (0.447 mol) of PLURIOL® E-200 divinyl ether and 91.43 g (0.498 mol) of DMDO were mixed with 1.83 g (0.013 mol) of 1,2,3-propanethiol (PTT) and allowed to react exothermically for 72 hours. The mixture was then heated to  $80^\circ\text{C}$ . To the heated reaction mixture was added 0.2 g (1 mmol) of VAZO® 67. The reaction mixture was maintained at  $80^\circ\text{C}$ ., and the reaction proceeded substantially to completion after 3 hours to afford 200 g (0.06 mol, yield 100%) of a polymer having a  $T_g$  of  $-66^\circ\text{C}$ . and a viscosity of 55 poise.

## Example 6

In a small jar, 14.0 g (0.055 mol) of PLURIOL® E-200 divinyl ether, 6.16 g (0.0336 mol) of DMDO and 5.38 g (0.0336 mol) of DMDS were mixed with 0.42 g (0.017 mol) of TAC (briefly heated to melt the TAC) and heated to  $82^\circ\text{C}$ . To the heated reaction mixture was added 0.2 g (0.001 mol) of VAZO® 67. The reaction proceeded substantially to completion after 18 hours to afford 26 g (8.4 mmol, yield 100%) of a polymer having a  $T_g$  of  $-63^\circ\text{C}$ . and a viscosity of 80 poise.

## Example 7

In a small jar, 13.55 g (0.054 mol) of PLURIOL® E-200 divinyl ether, 10.44 g (0.057 mol) of DMDO and 1.44 g (8.1 mmol) of ethylcyclohexanedithiol (ECHDT) were mixed with 0.40 g (1.6 mmol) of TAC (heated briefly to melt the TAC) and heated to  $82^\circ\text{C}$ . To the heated reaction mixture was added 0.2 g (0.001 mol) of VAZO® 67. The reaction proceeded substantially to completion after 5 hours to afford 26 g (8.1 mmol, yield 100%) of a polymer having a  $T_g$  of  $-66^\circ\text{C}$ . and a viscosity of 58 poise.

## Example 8

In a small glass jar, 9.11 g (0.036 mol) of PLURIOL® E-200 divinyl ether, 5.71 g (0.031 mol) of DMDO, 1.52 g (7.8 mmol) of ECHDT, 5.08 g (0.031 mol) of DMDS and 4.11 g (0.024 mol) of hexanediol divinyl ether (HD-DVE) were mixed with 0.39 g (1.6 mmol) of TAC (heated briefly to dissolve the TAC) and heated to  $82^\circ\text{C}$ . To the heated reaction mixture was added 0.6 g (3.1 mmol) of VAZO® 67. The reaction proceeded substantially to completion after about 45 hours to afford 2.6 g (7.8 mmol, yield 100%) of a polymer having a  $T_g$  of  $-66^\circ\text{C}$ . and a viscosity of 304 poise. The polymer had a cloudy appearance.

Each of the foregoing polymers was evaluated for odor. The following scale was employed: 3: strong, offensive odor; 2: moderate odor; 1: slight odor; 0: substantially odorless.

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The polymer described in Example 3 of U.S. Pat. No. 4,366,307 was used as a control. This polymer (the "control polymer") had an odor of 3.

Results were as follows:

Polymer	Odor
1	1
2	1
3	3
4	1
5	1
6	1
7	1
8	2

All of the liquid polythioethers thus had little or moderate odor except polymer 3, which had a strong odor.

The polymers prepared in Examples 1–8 were then cured. Curing was carried out using the uncompound resins with a curing agent and DABCO accelerator. The curing agent had the following composition:

epoxy novolac <sup>1</sup> (equivalent weight 175.5)	22 wt %
hydantoin epoxy <sup>2</sup> (equivalent weight 132)	34 wt %
calcium carbonate	34 wt %
carbon black	5 wt %
silane adhesive promoter	5 wt %

<sup>1</sup>DEN-431 epoxy novolac available from Dow Chemical of Midland, Michigan.

<sup>2</sup>ARACAST XU A4 238 hydantoin epoxy available from Ciba-Geigy.

The cured resins were evaluated for odor according to the procedure set forth above. The  $T_g$  and the percent weight gain after immersion in JRF type 1 for one week at room temperature and pressure were also measured for each of the cured resins. The volume swell and weight gain percentages were determined for each cured material as follows:

$w_1$  = initial weight in air

$w_2$  = initial weight in  $\text{H}_2\text{O}$

$w_3$  = final weight in air

$w_4$  = final weight in  $\text{H}_2\text{O}$

% volume swell =  $100 \times [(w_2 + w_3) - (w_1 + w_4)] / (w_1 - w_2)$

% weight gain =  $100 \times (w_3 - w_1) / w_1$

The results are given in Table 1:

TABLE 1

Cured Resin	1	2	3	4	5	6	7	8
Odor	0	0	0	0	0	0	0	0
$T_g$ ( $^\circ\text{C}$ )	-59	-61	-61	-63	-62	-56	-59	-58
% fuel swell	19	22	—	—	23	19	24	27
% wt. Gain	14	15	15	23	15	15	19	20

In comparison, the control polymer had an odor of 1–2 when cured.

## Example 9

Polythioethers having a number average molecular weight of 2100 and an average SH functionality F of 2.1 were prepared by combining a divinyl ether with a dithiol as shown in Table 2 and reacting the materials as previously described herein. The uncompound polythioethers were then cured using 15 g of the curing agent described above and 0.30 g of DABCO. For each polythioether so prepared,

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the following quantities were measured: viscosity (uncured material, poise p); Shore A hardness (cured material, Rex durometer value); % weight gain (cured material) after one week at 140° F. (60° C.) and atmospheric pressure in JRF type 1; and  $T_g$  (cured material, °C.). Results were as follows:

TABLE 2

Dithiol Divinyl Ether	ECHDT	DMDS	DMDO	HDT <sup>a</sup>
DEG-DVE	145 p 44 Rex 27% -53	(solid) 94 Rex 3% -63	27 p 25 Rex 14% -69	24 p 25 Rex 29% -77
PLURIOL® <sup>b</sup>	77 p 43 Rex 27% -57	41 p 47 Rex 11% -61	59 p 27 Rex 18% -67	25 p 23 Rex 30% -76
BD-DVE <sup>c</sup>	185 p 42 Rex 44% -59	(solid) — — —	(solid) 20 Rex 21% -79	(solid) 22 Rex 44% -85
HD-DVE	155 p 50 Rex 57% -60	(solid) — — -63	(solid) 14 Rex 27% -78	(soft solid) 29 Rex 68% -86
Poly-THF <sup>c</sup>	91 p 30 Rex 64% -69	(solid) 75 Rex 29% -79	27 p 17 Rex 37% -79	— — — —

<sup>a</sup>PLURIOL® E200 divinyl ether

<sup>b</sup>Butanediol divinyl ether

<sup>c</sup>polytetrahydrofuran divinyl ether

<sup>d</sup>Hexanediol

From the foregoing table it is apparent that the following combinations of divinyl ether and dithiol afford liquid polythioethers having good fuel resistance and low temperature flexibility when cured: PLURIOL® E-200/DMDO; and DEG-DVE/DMDO. Other potentially useful combinations include DEG-DVE/ECHDT; DEG-DVE/HDT; PLURIOL® E-200/ECHDT; PLURIOL® E-200/HDT; and poly-THF/DMDO. PLURIOL® E-200/DMDS also has excellent fuel resistance and low temperature flexibility when cured, but the uncompounded material does not remain in a liquid state for an extended period of time.

## Example 10

## Addition of DMDS to PLURIOL®/DMDO Polymers

Four liquid polythiol ether polymers were prepared as previously described herein. The polymers had the following compositions (listed values are molar equivalents):

	1	2	3	4
PLURIOL® E-200	6.6	6.6	6.6	6.6
DMDO	8	6	4.5	4
DMDS	0	2	3.5	4

To each of these polymers was added 0.2 molar equivalents of TAC to afford a number average molecular weight of about 3000 and a SH functionality F of 2.2. Each resultant uncompounded polymer was cured as in Example 9 (15 g of the curing agent composition and 0.30 g of DABCO). For each polymer, the following properties were measured:  $T_g$  (resin, °C.);  $T_g$  (cured, °C.); viscosity (p); % swell in JRF type 1; % weight gain in JRF type 1; and % weight gain in water. Results are given in Table 3.

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TABLE 3

	1	2	3	4
$T_g$ (resin)	-67	-66	-64	-63
$T_g$ (cured)	-59	-58	-56	-56
Viscosity JRF	59	53	62	80
% Swell	24	21	21	20
% Wt Gain	18	15	16	16
H <sub>2</sub> O	11.8	11.5	7.4	7.5
% Wt Gain				

All of the foregoing polymers displayed excellent fuel resistance. Polymers 1 and 2 in particular also displayed excellent low temperature flexibility when tested according to AMS 3267 § 4.5.4.7.

## Example 11

## Addition of ECHDT to PLURIOL®/DMDO Polymers

Four liquid polythiol ether polymers were prepared as previously described herein. The polymers had the following compositions (listed values are molar equivalents):

	1	2	3	4
PLURIOL® E-200	6.6	6.6	6.6	6.6
DMDO	8	7	6	5
ECHDT	0	1	2	3

Each of these polymers had a number average molecular weight of about 3000 and a SH functionality F of 2.2. Each resultant uncompounded polymer was cured as in Example 10. For each polymer, the following properties were measured:  $T_g$  (resin, °C.);  $T_g$  (cured, °C.); viscosity (p); % swell in JRF type 1; % weight gain in JRF type 1; and % weight gain in water. Results are given in Table 4.

TABLE 4

	1	2	3	4
$T_g$ (resin)	-67	-66	-65	-64
$T_g$ (cured)	-59	-59	-58	-56
Viscosity JRF type 1	59	36	44	50
% Swell	24	21	28	29
% Wt Gain	18	18	19	19
H <sub>2</sub> O	11.8	10.8	8.3	7.8
% Wt Gain				

All of the foregoing polymers displayed good fuel resistance and low temperature flexibility.

## Example 12

In a 250 mL 3-neck flask equipped with a stirrer, thermometer and condenser, 87.7 g (0.554 mol) of DEG-DVE and 112.3 g (0.616 mol) of DMDO are mixed and heated to 77° C. (about 170° F.). To the mixture is added 0.8 g (4.2 mmol) of VAZO® 67 catalyst. The reaction mixture is reacted at 82° C. (about 180° F.) for about 6 hours to afford 200 g (0.06 mol, yield 100%) of a low viscosity liquid polythioether resin having a thiol equivalent weight of 1625 and a SH functionality F of 2.0.

## Example 13

In a 250 mL 3-neck flask equipped with a stirrer, thermometer and condenser, 26.7 g (0.107 mol) of TAC, 56.4 g

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(0.357 mol) of DEG-DVE and 117.0 g (0.642 mol) of DMDO are mixed and heated to 77° C. (about 1700° F.). To the mixture is added 0.8 g (4.2 mmol) of VAZO 67 catalyst. The reaction mixture is reacted at 82° C. (about 180° F.) for about 6 hours to afford 200 g (0.07 mol, yield 100%) of a high viscosity liquid polythioether resin having an equivalent weight of 800 and a SH functionality F of about 3.5.

## Example 14

## Sealant Composition

A sealant composition including the DMDO/DEG-DVE polythioether polymer of Example 1 was compounded as follows (amounts in parts by weight):

DMDO/DEG-DVE Polythioether	100
Calcium carbonate	60
Magnesium oxide	1
Phenolic resin <sup>3</sup>	1
DMP-30	1
Isopropyl alcohol	3

The compounded polymer was mixed intimately with the epoxy resin curing agent of Examples 9–11 above, in the weight ratio of 10:1 and cured at ambient temperature and humidity. Tensile strength and elongation were evaluated according to ASTM 3269 and AMS 3276. The die used to prepare the test samples is described in ASTM D 412. The die used to prepare test samples for tear strength testing is described in ASTM D1004. The following physical properties were obtained for the cured composition:

Cure hardness at 25° C.	60 Shore A
Tensile strength at break	550 psi
Elongation at break	600%
Notched tear strength	100 p/i
Low-temperature flexibility	Passed
(AMS 3267 §4.5.4.7)	

## Example 15

## Sealant Composition

A sealant composition including the ECHDT/DEG-DVE polythioether polymer of Example 9 was compounded as follows (amounts in parts by weight):

ECHDT/DEG-DVE Polythioether	100
Calcium carbonate	54
Hydrated aluminum oxide	20
Magnesium oxide	1
Phenolic resin of Example 14	1
Hydrogenated terphenyl plasticizer	6
DMP-30	1
Isopropyl alcohol	3

The compounded polymer was mixed intimately with an epoxy resin curing agent of Examples 9–12 above in the weight ratio of 10:1 and cured at ambient temperature and humidity. The following physical properties were obtained for the cured composition:

Cure hardness at 25° C.	72 Shore A
Tensile strength at break	550 psi

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-continued

Elongation at break	450%
Notched tear strength	85 p/i
Low-temperature flexibility	Passed

## Example 16

## OH-Terminated Capped Polythioether

In a 500 ml flask, 275.9 g (1.09 mol) PLURIOL® E-200 divinyl ether, 174.7 g (0.95 mol) DMDO, 28.7 g (0.30 mol) 3-mercaptopropanol and 1.83 g (7.3 mmol) TAC were mixed. The mixture was heated to 70° C., and 2.3 g (12 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85–90° C. for 4 hours to afford 480 g (0.15 mol, yield 100%) of a polymer having an OH equivalent weight of 1670 (number average molecular weight=3200, OH functionality F=2.05).

## Example 17

## OH-Terminated Capped Polythioether

In a 250 ml flask, 104.72 g (0.57 mol) DMDO, 80.73 g (0.51 mol) DEG-DVE and 14.96 g (0.13 mol) butanediol monovinyl ether were mixed and heated to 75° C. To the heated mixture 0.60 g (3 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 75–85° C. for 6 hours to afford 200 g (0.064 mol, yield 100%) of a clear, nearly colorless polymer with very low odor and a viscosity of 79 poise at 20° C. The OH equivalent weight was 1570 (number average molecular weight=3200, OH functionality F=2.00).

## Example 18

## Sealant Composition

A sealant composition including the DMDO/DEG-DVE polythioether polymer of Example 1 was compounded as follows (amounts in weight percent):

## Base Composition

Wt. %	Component
0.277	Tung oil
1.053	triethylene diamine, diazabicyclo (2,2,2) octane <sup>4</sup>
55.466	Polythioether polymer of Example 1
0.831	Phenolic resin adhesion promoter <sup>5</sup>
0.554	Phenolic/polysulfide adhesion promoter <sup>6</sup>
0.277	Titanate <sup>7</sup>
1.108	Amino functional silane <sup>8</sup>
0.222	Fumed silica
0.554	Titanium dioxide
0.831	Amorphous silica
8.320	Aluminum hydroxide
30.506	precipitated calcium carbonate

<sup>4</sup>DABCO triethyl amine, diazabicyclo (2,2,2) octane available from Air Products & Chemicals.

<sup>5</sup>METHYLON 75108 phenolic resin available from Occidental Chemical.

<sup>6</sup>The phenolic/polysulfide adhesion promoter was prepared by reacting about 31% VARCUM 29202 phenolic resin, 66% Thiokol LP-3 polysulfide and 3% of a polymer prepared according to Example 4 of U.S. Pat. No. 4,623,711 (at a ratio of 1 mole diol to 1 mole polysulfide) (incorporated by reference herein) at a temperature of about 150° F. (65° C.) for 45 mins, then heated to 230° F. (110° C.) over a 45–60 minute period, then heated at 230° F. (110° C.) for 165 mins.

<sup>7</sup>TYZOR TBT titanate available from E.I. duPont de Nemours Company.

<sup>8</sup>A-1100 amino functional silane available from OSI Specialties, Inc.

## Accelerator

Wt. %	Component
26.525	Bisphenol A diglycidyl ether <sup>9</sup>
17.684	Epoxy novolac <sup>10</sup>
10.699	Plasticizer <sup>11</sup>
42.440	Calcium carbonate
0.221	Carbon black
0.088	Carbamate salt <sup>12</sup>
2.247	Epoxy functional silane <sup>13</sup>
0.085	Deionized water
0.011	Diphenylguanidine

<sup>9</sup>EPON 828 bisphenol A diglycidyl ether available from Shell Chemical.

<sup>10</sup>DEN 431 epoxy novolac available from Dow Chemical.

<sup>11</sup>HB-40 plasticizer available from Monsanto Co.

<sup>12</sup>Ferbam 76% WDG carbamate salt available from Cabot Corp.

<sup>13</sup>A-187 epoxy functional silane available from OSi Specialties, Inc.

Therefore, we claim:

1. A sealant formulation prepared from components comprising:

- (a) at least one ungelled mercapto-terminated polymer prepared by reacting reactants comprising at least one polyvinyl ether monomer and at least one polythiol material;
- (b) at least one curing agent reactive with a mercapto group of the mercapto-terminated polymer; and
- (c) at least one additive selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

2. The sealant formulation of claim 1, wherein said mercapto-terminated polymer has a glass transition temperature of less than -50° C.

3. The sealant formulation of claim 1, wherein said mercapto-terminated polymer has a viscosity of less than about 500 poise at a temperature of about 25° C. and a pressure of about 760 mm Hg.

4. The sealant formulation of claim 1, wherein said mercapto-terminated polymer has a number average molecular weight of between 500 and 20,000.

5. The sealant formulation of claim 4, wherein said mercapto-terminated polymer has a number average molecular weight of between 2000 and 5000.

6. The sealant formulation of claim 1, wherein said mercapto-terminated polymer comprises 30 to 90 weight percent of the sealant formulation based upon total weight of the components used to prepare the sealant formulation.

7. The sealant formulation of claim 1, wherein said polyvinyl ether monomer is a divinyl ether monomer.

8. The sealant formulation of claim 7, wherein said divinyl ether monomer is selected from the group consisting of: divinyl ether, ethylene glycol divinyl ether, butanediol divinyl ether, hexanediol divinyl ether, diethylene glycol divinyl ether, triethylene glycol divinyl ether, trimethylolpropane trivinyl ether, tetraethylene glycol divinyl ether, cyclohexanedimethanol divinyl ether, polytetrahydrofuryl divinyl ether and mixtures thereof.

9. The sealant formulation of claim 1, wherein said polyvinyl ether monomer is a mixture of components comprising divinyl ether and polyvinyl ether having at least three vinyl groups.

10. The sealant formulation of claim 1, wherein said polyvinyl ether monomer comprises 20 to 60 mole percent of the reactants used to prepare the mercapto-terminated polymer.

11. The sealant formulation of claim 1, wherein said polyvinyl ether monomer has a pendant group selected from

the group consisting of alkyl groups, hydroxyl groups, alkoxy groups and mixtures thereof.

12. The sealant formulation of claim 1, wherein said polythiol material is a dithiol.

13. The sealant formulation of claim 12, wherein said dithiol material is selected from the group consisting of 1,2-ethanedithiol, 1,2-propanedithiol, 1,3-propanedithiol, 1,3-butanedithiol, 1,4-butanedithiol, 2,3-butanedithiol, 1,3-pentanedithiol, 1,5-pentanedithiol, 1,6-hexanedithiol, 1,3-dimercapto-3-methylbutane, dipentenedimercaptan, ethylcyclohexyldithiol, dimercaptodiethylsulfide, methyl-substituted dimercaptodiethylsulfide, dimethyl-substituted dimercaptodiethylsulfide, dimercaptodioxaoctane, 1,5-dimercapto-3-oxapentane and mixtures thereof.

14. The sealant formulation of claim 1, wherein said polythiol material has a pendant group selected from the group consisting of alkyl groups, hydroxyl groups, and alkoxy groups.

15. The sealant formulation of claim 1, wherein said polythiol material has a number average molecular weight ranging from 200 to 5000.

16. The sealant formulation of claim 1, wherein said polythiol material comprises from 40 to 80 mole percent of the reactants used to prepare the mercapto-terminated polymer.

17. The sealant formulation of claim 1, wherein said reactants from which said mercapto-terminated polymer are prepared further comprise a catalyst.

18. The sealant formulation of claim 1, wherein said curing agent is selected from a group consisting of polyolefins, polyacrylates, metal oxides, polyepoxides and mixtures thereof.

19. A sealant formulation prepared from components comprising:

- (a) at least one ungelled mercapto-terminated polymer prepared from reactants comprising diethylene glycol divinyl ether and dimercapto dioxaoctane;
- (b) at least one curing agent reactive with a mercapto group of the mercapto-terminated polymer; and
- (c) at least one additive selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

20. An aerospace sealant formulation prepared from components comprising:

- (a) at least one ungelled mercapto-terminated polymer prepared by reacting reactants comprising at least one polyvinyl ether monomer and at least one polythiol material;
- (b) at least one curing agent reactive with a mercapto group of the mercapto-terminated polymer; and
- (c) at least one additive selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

21. An electrical potting formulation prepared from components comprising:

- (a) at least one ungelled mercapto-terminated polymer prepared by reacting reactants comprising at least one polyvinyl ether monomer and at least one polythiol material;
- (b) at least one curing agent reactive with a mercapto group of the mercapto-terminated polymer; and
- (c) at least one additive selected from the group consisting of fillers, adhesion promoters, plasticizers and catalysts.

\* \* \* \* \*

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UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

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JONATHAN D. **ZOOK**, DAVID W. JORDAN, DEAN M. WILLARD,  
GEORGE JONES, AND MICHAEL COSMAN,  
Junior Party ("PBT")  
(U.S. Patents 6,525,168 and 6,723,827),

v.

JONATHAN D. **ZOOK**, SUSAN E. DEMOSS,  
DAVID W. JORDAN AND CHANDRA B. RAO  
Senior Party ("PRC")  
(U.S. Application 10/644,389).

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Patent Interference No. 105,555 (MPT)  
(Technology Center 1700)

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DECLARATION OF WILLIAM R. LAMBERT  
ZOOK EXHIBIT 1022

**ZOOK EXHIBIT 1022**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

I, William R. Lambert, declare that:

1. I am a patent attorney registered to practice before the U.S. Patent and Trademark Office (USPTO). My registration number is 44,857.
2. I am also an active member of the State Bar of California.
3. I am currently employed as a patent attorney by XenoPort, Inc., 3410 Central Expressway, Santa Clara, CA 95051.
4. Prior to my employment by XenoPort, Inc., I was employed as a patent attorney from about August 2002 to January 2005 by the law firm of Finnegan, Henderson, Farabow, Garrett and Dunner (Finnegan) in their California office now located at 3300 Hillview Drive, Palo Alto, CA 94304.
5. During the time I was employed by Finnegan I was responsible for a number of patent matters for the firm client PRC-DeSoto International, Inc. (formerly Courtaulds Aerospace, Inc.), a wholly-owned subsidiary of PPG Industries.
6. The client contact for PRC patent matters was William J. Uhl, a registered patent attorney (Registration No. 25,567), with whom I worked closely.
7. Among the matters I worked on with Mr. Uhl were U.S. Application No. 10/302,177, filed, prior to my involvement with the case, on November 21, 2002, and U.S. Application No. 10/644,389, filed August 19, 2003. These applications are part of a chain of divisional, continuation, and continuation-in-part applications, the relationship of which is depicted on Zook Exhibit 1003. The various relevant applications and

patents in the chain of applications referred to in Zook Exhibit 1003 are numbered 1 (for the application having the earliest filing date) to 6 (for the most recently filed application). For convenience, the various applications and patents will be referred to herein by both the application or patent number and by the sequential numbers appearing in Zook Exhibit 1003.

8. I also worked with Mr. Uhl with regard to filing a reissue application of U.S. Patent No. 5,912,319 (No. 1). The reissue application, Application No. 10/788,965, was filed on February 25, 2004, along with a request for interference with U.S. Patent Nos. 6,525,168 and 6,723,827 (respectively, the '168 and '827 patents).

9. At some point after February 25, 2003 (the issuance date of the '168 patent), and August 19, 2003, Mr. Uhl commenced discussions with me and others regarding the possibilities of provoking an interference with the '168 patent.

10. Application No. 10/644,389 (No. 6 on Zook Exhibit 1003) was filed by me on August 19, 2003 in contemplation of being used as a vehicle for provoking such an interference. It was also contemplated that a reissue of U.S. Patent No. 5,912,319 (No. 1) could be used for this purpose.

11. At some point prior to February 25, 2004 (one year from the issue date of the '168 patent and also the filing date of the reissue of U.S. Patent No. 5,912,319 (No. 1)), it was decided to request an interference with the '168 patent in both the reissue application and Application No. 10/644,389 (No. 6). Accordingly, I prepared an application for reissue of U.S. Patent No. 5,912,319 (No. 1), a request for interference in the reissue application, and a request for interference in the previously filed Application

No. 10/644,389 (No. 6). All of these papers were filed by me in the USPTO on February 25, 2004.

12. I have been advised that Application No. 10/644,389 (No. 6) has been placed in interference with the '168 and '187 patents. I have also been advised that there appears to be a break in continuity between Application No. 10/302,177 (No. 5) and 10/644,389 (No. 6).

### **THE BREAK IN CONTINUITY**

13. In Application No. 10/302,177 (No. 5), a non-final Office Action with a shortened three-month period for response was mailed on February 19, 2003. Zook Exhibit 1004. Neither a response nor a petition for extension of time for reply to this Office Action was filed in that case.

14. Application No. 10/644,389 (No. 6) was filed on August 19, 2003, the last day that a response could have been filed in Application No. 10/302,177 (No. 5) had a petition for extension of time, including the appropriate fee, been filed in that case.

15. Failure to file a petition for extension of time in Application No. 10/302,177 (No. 5) was an inadvertent oversight. Thus, the premature abandonment of that application without preserving continuity with Application No. 10/644,389 (No. 6) was unintentional.

16. That the abandonment of Application No. 10/302,177 (No. 5) without preserving continuity with Application No. 10/644,389 (No. 6) was unintentional, and that it was my intent and the intent of PRC to preserve continuity, is in part evidenced by the fact that Application No. 10/644,389 (No. 6) was filed on the last day that copendency could

have been preserved by filing a petition for extension of time in Application No.

10/302,177 (No. 5). There would be no reason to file the application on this particular day if copendency was not desired.

17. Moreover, in the transmittal letter (Zook Exhibit 1006) submitted with Application No. 10/644,389 (No. 6), that application was referred to as "a Continuation Application" of "pending prior Application No. 10/302,177." Specifically, the first sentence in the body of the transmittal letter provides in part:

This is a request for filing a Continuation Application under  
37 C.F.R. § 1.53(b) of pending prior Application No.  
10/302,177, filed on November 21, 2001 . . . .

Application No. 10/644,389 (No. 6) would not have been referred to as a "Continuation," and Application No. 10/302,177 (No. 5) would not have been referred to as "pending" if I did not believe Application No. 10/302,177 (No. 5) was pending when Application No. 10/644,389 (No. 6) was filed. Thus, I clearly intended to preserve, and believed I had preserved, copendency.

18. In addition, in paragraph 9 on page 2 of the transmittal letter (Zook Exhibit 1006), the first paragraph of the specification of the newly filed Application No. 10/644,389 (No. 6) was amended to recite a claim to priority to Application No. 10/302,177 (No. 5) in accordance with 37 C.F.R. § 1.78(a)(2). This amendment would not have been done had I not intended to preserve copendency with the prior application.

19. Although a Notice of Abandonment (Zook Exhibit 1005) was received in Application No. 10/302,177 (No. 5), it does not indicate when that application went abandoned, therefore I was unaware it was prematurely abandoned.

20. Furthermore, Application No. 10/644,389 (No. 6) was filed for the purpose of provoking an interference with the '168 patent. Clearly, it would be illogical to file that application for purposes of provoking an interference while intentionally breaking continuity with the chain of earlier cases and thus losing the right to rely on these cases for priority purposes. This is especially self evident given the earliest priority date (February 19, 1997) available for Application No. 10/644,389 (No. 6) compared to the earliest possible priority date available for the '168 patent (March 9, 2000).

21. The Filing Receipt (Zook Exhibit 1011) for Application No. 10/644,389 (No. 6) dated November 17, 2003. The Filing Receipt contained the following notations:

Domestic Priority data as claimed by applicant

This application is a CON of 10/302,177 11/21/2002 ABN\*  
which is a CON of 09/756,573 of 01/08/2001 PAT  
6,509,418\*  
which is a CIP of 08/928,972 09/12/1997 PAT 6,172,179  
which is a CIP of 08/802,130 02/19/1997 PAT 5,912,319  
This application 10/644,389  
is a CIP of 09/318,500 05/25/1999 PAT 6,232,401  
which is a DIV of 08/802,130 02/19/1997 PAT 5,912,319  
and said 09/756,573 01/08/2001  
claims benefit of 60/182,396 02/14/2000  
and claims benefit of 60/215,548 06/30/2000  
(\*)Data provided by applicant is not consistent with PTO  
records

The asterisks after the first two list applications appeared to me to be in error, particularly since the Filing Receipt did not indicate any problem with the priority claim to applications filed prior to Application No. 09/756,573 (No. 4). Certainly nothing in the notations appearing on the filing receipt indicated to me there was any problem with copendency between Application No. 10/302,177 (No. 5) and Application No. 10/644,389 (No. 6).

22. I requested a corrected filing receipt (Zook Exhibit 1012), again reflecting my belief that Application No. 10/644,389 (No. 6) was entitled to claim priority from Application No. 10/302,177 (No. 5) and that Application No. 10/302,177 (No. 5) was not prematurely abandoned. Moreover, the request for a corrected Filing Receipt was granted. Zook Exhibit 1013.

23. Subsequent to submitting the request for a corrected Filing Receipt, I left the Finnegan Firm and had no further involvement with the prosecution of Application No. 10/644,389 (No. 6).

24. Thus, nothing that happened subsequent to filing Application No. 10/644,389 (No. 6) alerted me to the break in copendency. To the best of my knowledge, neither I nor anyone affiliated with PRC ever intended to abandon Application No. 10/302,177 (No. 5) without preserving copendency, or was aware of the inadvertent premature abandonment of that Application until the break in continuity was discovered by interference counsel after the interference was declared. Thus, the abandonment and delay in filing a petition for extension of time in Application No. 10/302,177 (No. 5) has remained unintentional at all times.

#### **THE INCORRECT CLAIM TO THE ALTERNATIVE CHAIN OF PRIORITY APPLICATIONS**

25. In addition to the chain of priority cases discussed above, Application No. 10/644,389 (No. 6) includes an alternative priority chain that involves Application No. 09/318,500 (No. 3).

26. Specifically, the main chain of priority applications begins with Application No. 08/802,130 (No. 1), and runs through Application Nos. 08/928,972 (No. 2), 09/756,573 (No. 4), and 10/302,177 (No. 5) to the application currently involved in the interference, Application No. 10/644,389 (No. 6). The alternate chain of priority applications also begins with Application No. 08/802,130 (No. 1) and runs through Application Nos. 09/318,500 (No. 3), 09/756,573 (No. 4), and 10/302,177 (No. 5) to Application No. 10/644,389 (No. 6). See Zook Exhibit 1003 (in the exhibit, the correct chains of priority cases are connected by solid lines).

27. Application No. 09/756,573 (No. 4) (now U.S. Patent No. 6,509,418), the first application in the relevant chain of applications that could have claimed priority to Application No. 09/318,500 (No. 3), properly recited both the alternative chain that includes Application No. 09/318,500 (No. 3) and the chain that does not. Specifically, in the alternative priority chain, as set forth in Application No. 09/756,573 (No. 4), that application is properly identified as a continuation-in-part of Application No. 09/318,500 (No. 3), which application is a divisional of Application No. 08/802,130 (No. 1). See Zook Exhibit 1014, first paragraph of Col 1.

28. Application No. 10/302,177 (No. 5) was filed prior to my involvement with the case. At the time that application was filed, the wording of the alternative priority claim was changed only slightly from what it had been in Application No. 09/756,573 (No. 4). See Zook Exhibits 1014 and 1007, paragraph 9, page 2. Because the wording was changed only slightly, the alternative priority claim was ambiguous and it was not clear Application No. 09/318,500 (No. 3) is a parent continuation-in-part of Application No. 09/756,573 (No. 4). Rather, Application No. 10/318,500 (No. 3) appears to be a parent

continuation-in-part of Application No. 10/302,177 (No. 5). Application No. 10/302,177 (No. 5) could not directly claim priority from Application No. 09/318,500 (No. 3) because the two applications were never copending. In contrast, Application No. 09/318,500 (No. 3) and Application No. 09/756,573 (No. 4) were copending and the latter application could properly claim priority from the former application.

29. Subsequently, I became involved with the prosecution of Application No. 10/302,177 (No. 5), and noticed that the priority information set forth on the filing receipt did not correspond to that set forth in the transmittal letter. Compare Zook Exhibits 1021 and 1007, paragraph 9, page 2. I filed a Request for a Corrected Filing Receipt (Zook Exhibit 1015) to correct the inconsistency so that the priority information corresponded to the priority information set forth in the transmittal letter (Zook Exhibit 1007, paragraph 9, page 2). At the time, I did not notice the ambiguity.

30. Subsequently, Application No. 10/644,389 (No. 6) was filed by me, and Application No. 10/302,177 (No. 5) was abandoned. At the time of filing Application No. 10/644,389 (No. 6), I made some punctuation changes in the priority claim as recited in Application No. 10/302,177 (No. 5) and inserted a reference to that application, but otherwise did not change the alternate priority claim from what had been previously recited. Compare Zook Exhibits 1006, at paragraph 9, page 2, and 1007, at paragraph 9, page 2.

31. At the time of filing Application No. 10/644,389 (No. 6), I did not realize that the priority claim to the alternative chain of applications could be construed to incorrectly suggest that Application No. 10/644,389 (No. 6) claimed priority directly from Application

No. 09/318,500 (No. 3) (the dash line on Zook Exhibit 1003). However, because these two applications were not copending, priority to Application 09/318,500 (No. 3) could only be obtained through a chain of continuation and continuation-in-part applications including Application No. 09/756,573 (No. 4).

32. The ambiguous or improper alternative priority claims involving Application No. 09/318,500 (No. 3) were made by inadvertent unintentional oversight. It was my intent at all times that there be a properly claimed alternative priority chain that included Application No. 09/318,500 (No. 3).

33. As noted above in paragraphs 21 and 22, I requested an amended Filing Receipt in Application No. 10/644,389 (No. 6) because the original Filing Receipt contained some erroneous notations. At that time, I did not appreciate any deficiency in the alternative priority chain.

34. Moreover, at no time was I, nor to the best of my knowledge was anyone else affiliated with PRC, aware of any deficiency in the priority claims involving Application No. 09/318,500 (No. 3) until the defect in the alternative chain was discovered by interference counsel after the interference was declared. Thus, the entire delay in making the correct priority chain involving Application 09/318,500 (No. 3) has remained unintentional at all times.

35. The correct alternative priority claim in both Application No. 10/302,177 (No. 5) and Application No. 10/644,389 (No. 6) should recite "Said Application No. 09/756,573 (No. 4) is also a continuation-in-part of Application No. 09/318,500 (No. 3), which is a divisional of Application No. 08/802,130 (No. 1)." Specifically, Application No.

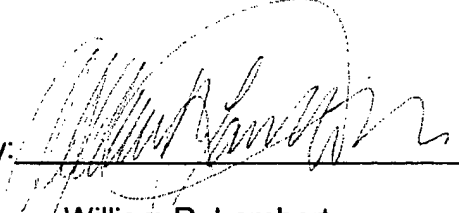
09/756,573 (No. 4) is a continuation in-part of both Application No. 08/928,972 (No. 2) and Application No. 09/318,500 (No. 3).

36. That this error was inadvertent, unintentional and made in good faith is evident in that it does not affect entitlement to benefit of the filing date of the earliest application in the chain, Application No. 08/802,130 (No. 1).

37. In signing this Declaration, I understand that the Declaration will be filed as evidence in a contested case before the Board of Patent Appeals and Interferences of the United States Patent and Trademark Office. I acknowledge that I may be subject to cross examination in the case and that cross examination will take place within the United States. If cross examination is required of me, I will appear for cross examination within the United States during the time allotted for cross examination.

38. The undersigned further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing therefrom.

Date: October 1, 2007

By:   
William R. Lambert

Filed on behalf of: Senior Party Jonathan D. Zook

By: Jerry D. Voight  
Wesley B. Derrick  
FINNEGAN, HENDERSON, FARABOW,  
GARRETT & DUNNER, L.L.P.  
3300 Hillview Avenue  
Palo Alto, California 94304  
Telephone: 650.849.6600  
Facsimile: 650.849.6666

UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

---

JONATHAN D. **ZOOK**, DAVID W. JORDAN, DEAN M. WILLARD,  
GEORGE JONES, AND MICHAEL COSMAN,  
Junior Party ("PBT")  
(U.S. Patents 6,525,168 and 6,723,827),

v.

JONATHAN D. **ZOOK**, SUSAN E. DEMOSS,  
DAVID W. JORDAN AND CHANDRA B. RAO  
Senior Party ("PRC")  
(U.S. Application 10/644,389).

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Patent Interference No. 105,555 (MPT)  
(Technology Center 1700)

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**DECLARATION OF WILLIAM J. UHL**  
**ZOOK EXHIBIT 1023**

**ZOOK EXHIBIT 1023**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

I, William J. Uhl, declare that:

1. I am a patent attorney registered to practice before the U.S. Patent and Trademark Office (USPTO). My registration number is 25,567
2. I am also an active member of the State Bar of Pennsylvania.
3. Since 1971, I have been employed as a patent attorney by PPG Industries, Inc., One PPG Place, Pittsburgh, PA, 15272.
4. I have been responsible, *inter alia*, for patent matters for PRC-DeSoto International, Inc. (formerly Courtaulds Aerospace, Inc.), which is a wholly-owned subsidiary of PPG Industries, Inc., since about 1999.
5. During my employment as a patent attorney by PPG, I was involved in directing the prosecution of U.S. Application Nos. 08/802,130, 08/928,972, 09/318,500, 09/756,573, 10/302,177, and 10/644,389, and participated in the prosecution of these applications before the USPTO. These applications are part of a chain of divisional, continuation, and continuation-in-part applications, the relationship of which is depicted in Zook Exhibit 1003. The various relevant applications and patents in the chain of applications referred to in Zook Exhibit 1003 are numbered 1 (for the application having the earliest filing date) to 6 (for the most recently filed application). For convenience, the various applications and patents will be referred to herein by both the application or patent number and by the sequential numbers appearing in Zook Exhibit 1003.
6. In the course of my involvement with the prosecution of U.S. Application No. 10/302,177 (No. 5), filed on November 21, 2002, and U.S. Application No. 10/644,389

(No. 6), filed August 19, 2003, I worked with Mr. William R. Lambert, a patent attorney employed by the law firm of Finnegan, Henderson, Farabow, Garrett and Dunner (Finnegan).

7. I also worked with Mr. Lambert with regard to filing a reissue application of U.S. Patent No. 5,912,319 (No. 1). The reissue application, Application No. 10/788,965, was filed on February 25, 2004, along with a request for interference with U.S. Patent Nos. 6,525,168 and 6,723,827 (respectively, the '168 and '827 patents).

8. I also worked with Mr. Phil Makrogiannis, a patent attorney employed by Finnegan, in the filing of Application No. 10/302,177 (No. 5), which application was filed with a preliminary amendment and request for interference on November 21, 2002. Zook Exhibit 1016.

9. The request for interference filed with Application No. 10/302,177 (No. 5) was intended to provoke an interference with Application Nos. 09/802,427 and 10/368,135, now the involved '168 and '827 patents, respectively.

10. At some point after February 25, 2003 (the issuance date of the '168 patent), and August 19, 2003, I commenced discussions with Mr. Lambert and others regarding the possibilities of provoking an interference with the '168 patent.

11. Application No. 10/644,389 (No. 6 on Zook Exhibit 1003) was filed by Mr. Lambert at my direction on August 19, 2003, in contemplation of being used as a vehicle for provoking such an interference. It was also contemplated that a reissue of U.S. Patent No. 5,912,319 (No. 1) could be used for this purpose.

12. At some point prior to February 25, 2004 (one year from the issue date of the '168 patent and also the filing date of the reissue of U.S. Patent No. 5,912,319 (No. 1)), it was decided to request an interference with the '168 patent in both the reissue application and Application No. 10/644,389 (No. 6). Accordingly, I directed Mr. Lambert to prepare an application for reissue of U.S. Patent No. 5,912,319 (No. 1), a request for interference in the reissue application, and a request for interference in the previously filed Application No. 10/644,389 (No. 6). All of these papers were filed by Mr. Lambert in the USPTO on February 25, 2004.

13. I am aware that Application No. 10/644,389 (No. 6) has been placed in interference with the '168 and '187 patents. I have also been advised that there appears to be a break in continuity between Application Nos. 10/302,177 (No. 5) and 10/644,389 (No. 6).

#### **THE BREAK IN CONTINUITY**

14. In Application No. 10/302,177 (No. 5), a non-final Office Action with a shortened three-month period for response was mailed on February 19, 2003. Zook Exhibit 1004. Neither a response nor a petition for extension of time for reply to this Office Action was filed in that case.

15. Application No. 10/644,389 (No. 6) was filed on August 19, 2003, the last day that a response could have been filed in Application No. 10/302,177 (No. 5) had a petition for extension of time, including the appropriate fee, been filed in that case.

16. Failure to file a petition for extension of time in Application No. 10/302,177 (No. 5) was an inadvertent oversight. Thus, the premature abandonment of that application without preserving continuity with Application No. 10/644,389 (No. 6) was unintentional.

17. That the abandonment of Application No. 10/302,177 (No. 5) without preserving continuity with Application No. 10/644,389 (No. 6) was unintentional, and that it was my intent and the intent of PRC to preserve continuity, is in part evidenced by the fact that Application No. 10/644,389 (No. 6) was filed by Mr. Lambert on the last day that copendency could have been preserved by filing a petition for extension of time in Application No. 10/302,177 (No. 5). There would be no reason to file the application on this particular day if copendency was not desired.

18. Moreover, in the transmittal letter (Zook Exhibit 1006) submitted with Application No. 10/644,389 (No. 6), that application was referred to as "a Continuation Application" of "pending prior Application No. 10/302,177." Specifically, the first sentence in the body of the transmittal letter provides in part:

This is a request for filing a Continuation Application under  
37 C.F.R. § 1.53(b) of pending prior Application No.  
10/302,177, filed on November 21, 2001 . . . .

Application No. 10/644,389 (No. 6) would not have been referred to as a "Continuation," and Application No. 10/302,177 (No. 5) would not have been referred to as "pending" if Mr. Lambert and I did not believe Application No. 10/302,177 (No. 5) was pending when Application No. 10/644,389 (No. 6) was filed. Thus, we clearly intended to preserve, and believed we had preserved copendency.

19. In addition, in paragraph 9 on page 2 of the transmittal letter (Zook Exhibit 1006), the first paragraph of the specification of the newly filed Application No. 10/644,389 (No. 6) was amended to recite a claim to priority to Application No. 10/302,177 (No. 5) in accordance with 37 C.F.R. § 1.78(a)(2). This amendment would not have been done had Mr. Lambert and I not intended to preserve copendency with the prior application.

20. Although a Notice of Abandonment (Zook Exhibit 1005) was received in Application No. 10/302,177 (No. 5), it does not indicate when that application went abandoned, therefore I was unaware it was prematurely abandoned.

21. Furthermore, Application No. 10/644,389 (No. 6) was filed for the purpose of provoking an interference with the '168 patent. Clearly, it would be illogical to file that application for purposes of provoking an interference while intentionally breaking continuity with the chain of earlier cases and thus losing the right to rely on these cases for priority purposes. This is especially self evident given the earliest priority date (February 19, 1997) available for Application No. 10/644,389 (No. 6) compared to the earliest priority date available for the '168 patent (March 9, 2000).

22. The Filing Receipt (Zook Exhibit 1011) for Application No. 10/644,389 (No. 6) dated November 19, 2003, Request for Corrected Filing Receipt (Zook Exhibit 1012), and grant of the Request for Corrected Filing Receipt (Zook Exhibit 1013) indicated to me that the claim in Application No. 10/466,389 (No. 6) to priority from Application No. 10/302,177 (No. 5) was correct and proper.

23. After Mr. Lambert left the Finnegan firm, I assumed primary responsibility for prosecution of Application No. 10/644,389 (No. 6). On January 5, 2006, I participated in

an interview with Examiner Duc Truong and Finnegan attorneys Charles E. Van Horn and Jerry Voight where we discussed what claims of Application No. 10/466,389 (No. 6) would be entitled to the benefit of priority of Application No. 08/802,130 (No. 1). As recorded by Examiner Truong in the Interview Summary (Zook Exhibit 1017), certain claims were understood by all present to be entitled to the earlier filing date of Application No. 08/802,130, which necessarily requires that all present believed there was copendency between Application Nos. 10/302,177 (No. 5) and 10/644,389 (No. 6).

24. On January 13, 2006, an Associate Power of Attorney or Agent (Zook Exhibit 1018) was filed naming me as an Associate Attorney in Application No. 10/644,389 (No. 6), along with a Reply to Office Action (Zook Exhibit 1019) taking into account the Interview with Examiner Duc Truong. I believed that the amendment included in the Reply would place the claims in condition for a declaration of interference with U.S. Patent 6,525,168 and Application No. 10/368,135 (U.S. Patent 6,723,827).

26. On May 19, 2006, I filed a Request to Correct Inventorship in Application 10/644,389 (No. 6) in light of claim amendments directed to what I understood to be subject matter of the proposed interference with U.S. Patents 6,525,168 and 6,723,827.

27. On September 12, 2006, I signed a Response (Zook Exhibit 1020), which states that U.S. Patent Nos. 6,525,168 and 6,723,827 were not prior art to the claims of Application No. 10/644,389 (No. 6); this statement was made in the belief that there was no break in copendency.

28. Subsequent to the Response filed September 12, 2006, nothing substantive occurred in Application No. 10/644,389 (No. 6) until the Declaration of interference on July 10, 2007, and I was never alerted to any break in copendency.

29. To the best of my knowledge, neither I nor anyone affiliated with PRC ever intended to abandon Application No. 10/302,177 (No. 5) without preserving copendency, or was aware of the inadvertent premature abandonment of that Application until the break in continuity was discovered by interference counsel after the interference was declared. Thus, the abandonment and delay in filing a petition for extension of time in Application No. 10/302,177 (No. 5) has remained unintentional at all times.

**THE INCORRECT CLAIM TO THE  
ALTERNATIVE CHAIN OF PRIORITY APPLICATIONS**

30. In addition to the chain of priority cases discussed above, Application No. 10/644,389 (No. 6) includes an alternative priority chain that involves Application No. 09/318,500 (No. 3).

31. Specifically, the main chain of priority applications begins with Application No. 08/802,130 (No. 1), and runs through Application Nos. 08/928,972 (No. 2), 09/756,573 (No. 4), and 10/302,177 (No. 5) to the application currently involved in the interference, Application No. 10/644,389 (No. 6). The alternate chain of priority applications also begins with Application No. 08/802,130 (No. 1) and runs through Application Nos. 09/318,500 (No. 3), 09/756,573 (No. 4), and 10/302,177 (No. 5) to Application No. 10/644,389 (No. 6). See Zook Exhibit 1003 (in the exhibit, the correct chains of priority cases are connected by solid lines).

32. Application No. 09/756,573 (No. 4) (now U.S. Patent No. 6,509,418), the first application in the relevant chain of applications that could have claimed priority to Application No. 09/318,500 (No. 3), properly recited both the alternative chain that includes Application No. 09/318,500 (No. 3) and the chain that does not. Specifically, in the alternative priority chain, as set forth in Application No. 09/756,573 (No. 4), that application is properly identified as a continuation-in-part of Application No. 09/318,500 (No. 3), which application is a divisional of Application No. 08/802,130 (No. 1). See Zook Exhibit 1014, first paragraph of Col 1.

33. Application No. 10/302,177 (No. 5) was filed with a slight change to the wording of the alternative priority claim from what it had been in Application No. 09/756,573 (No. 4). See Zook Exhibits 1014 and 1007, paragraph 9, page 2. Because the wording was changed only slightly, the alternative priority claim was ambiguous and it was not clear Application No. 09/318,500 (No. 3) is a parent continuation-in-part of Application No. 09/756,573 (No. 4). Rather, Application No. 10/318,500 (No. 3) appears to be a parent continuation-in-part of Application No. 10/302,177 (No. 5). Application No. 10/302,177 (No. 5) could not directly claim priority from Application No. 09/318,500 (No. 3) because the two applications were never copending. In contrast, Application No. 09/318,500 (No. 3) and Application No. 09/756,573 (No. 4) were copending and the latter application could properly claim priority from the former application.

34. Subsequently, Mr. Lambert became involved with the prosecution of Application No. 10/302,177 (No. 5), and noticed that the priority information set forth on the filing receipt did not correspond to that set forth in the transmittal letter. Compare Zook Exhibits 1021 and 1007, paragraph 9, page 2. He filed a Request for a Corrected Filing

Receipt (Zook Exhibit 1015) to correct the inconsistency so that the priority information corresponded to the priority information set forth in the transmittal letter (Zook Exhibit 1007, paragraph 9, page 2). At the time, I did not notice the ambiguity.

35. Subsequently, Application No. 10/644,389 (No. 6) was filed by Mr. Lambert, and Application No. 10/302,177 (No. 5) was abandoned. At the time of filing Application No. 10/644,389 (No. 6), Mr. Lambert made some punctuation changes in the priority claim as recited in Application No. 10/302,177 (No. 5) and inserted a reference to that application, but otherwise did not change the alternate priority claim from what had been previously recited. Compare Zook Exhibits 1006, at paragraph 9, page 2, and 1007, at paragraph 9, page 2.

36. At the time of filing Application No. 10/644,389 (No. 6), I did not realize that the priority claim to the alternative chain of applications could be construed to incorrectly suggest that Application No. 10/644,389 (No. 6) claimed priority directly from Application No. 09/318,500 (No. 3) (the dash line on Zook Exhibit 1003). However, because these two applications were not copending, priority to Application 09/318,500 (No. 3) could only be obtained through a chain of continuation and continuation-in-part applications including Application No. 09/756,573 (No. 4).

37. The ambiguous or improper alternative priority claims involving Application No. 09/318,500 (No. 3) were made by inadvertent unintentional oversight. It was my intent at all times that there be a properly claimed alternative priority chain that included Application No. 09/318,500 (No. 3).

38. As noted above in paragraph 22, where Mr. Lambert obtained a corrected filing receipt for other reasons, I did not appreciate any deficiency in the alternative priority chain.

39. Moreover, at no time was I, nor to the best of my knowledge was anyone else affiliated with PRC, aware of any deficiency in the priority claims involving Application No. 09/318,500 (No. 3) until the defect in the alternative chain was discovered by interference counsel after the interference was declared. Thus, the entire delay in making the correct priority chain involving Application 09/318,500 (No. 3) has remained unintentional at all times.

40. The correct alternative priority claim in both Application No. 10/302,177 (No. 5) and Application No. 10/644,389 (No. 6) should recite "Said Application No. 09/756,573 (No. 4) is also a continuation-in-part of Application No. 09/318,500 (No. 3), which is a divisional of Application No. 08/802,130 (No. 1)." Specifically, Application No. 09/756,573 (No. 4) is a continuation in-part of both Application No. 08/928,972 (No. 2) and Application No. 09/318,500 (No. 3).

41. That this error was inadvertent, unintentional and made in good faith is evident in that it does not affect entitlement to benefit of the filing date of the earliest application in the chain, Application No. 08/802,130 (No. 1).

42. In signing this Declaration, I understand that the Declaration will be filed as evidence in a contested case before the Board of Patent Appeals and Interferences of the United States Patent and Trademark Office. I acknowledge that I may be subject to cross examination in the case and that cross examination will take place within the

United States. If cross examination is required of me, I will appear for cross examination within the United States during the time allotted for cross examination.

43. The undersigned further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing therefrom.

Date: 2 OCTOBER 2007

By: William J. Uhl

William J. Uhl



Application No. 09/756,573

PATENT  
Customer No. 22,852  
Attorney Docket No. 8303.0042-04

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: )  
Jonathan D. ZOOK et al. ) Group Art Unit: 1712  
Application No.: 09/756,573 ) Examiner: To Be Assigned  
Filed: January 8, 2001 )  
For: SEALANTS AND POTTING )  
FORMULATIONS INCLUDING )  
MERCAPTO-TERMINATED )  
POLYMERS PRODUCED BY )  
THE REACTION OF A )  
POLYTHIOL AND POLYVINYL )  
ETHER MONOMER )

Assistant Commissioner  
for Patents  
Washington, DC 20231

OFFICE OF INITIAL PATENT EXAMINATION

Sir:

**REQUEST FOR CORRECTED FILING RECEIPT**

We are forwarding herewith a copy of a Filing Receipt for the above-identified patent application. As indicated in ink on the attached copy, there are two errors in the Filing Receipt. The first error is the first name and middle initial of the second inventor; the name should be Susan E. DeMoss. The second error is under the Domestic Priority Data as claimed by Applicants. The text should read:

--This application is a CIP of 08/928,972 09/12/1997 PAT 6,172,179 which is a CIP of 08/802,130 02/19/97 PAT 5,912,319. This application is also a CIP of 09/318,500 05/25/1999 PAT 6,232,401 which is a DIV of 08/802,130 02/19/97 PAT

LAW OFFICES  
FINNEGAN, HENDERSON,  
FARABOW, GARRETT  
& DUNNER, L.L.P.  
STANFORD RESEARCH PARK  
700 HANSEN WAY  
PALO ALTO, CALIF. 94304  
650-649-6600

**ZOOK EXHIBIT 1026**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

5,912,319. This application 09/756,573 claims benefit of 60/215,548 06/30/2000 and claims benefit of 60/182,396 02/14/2000.—


It is respectfully requested that a corrected Filing Receipt be issued as soon as possible.

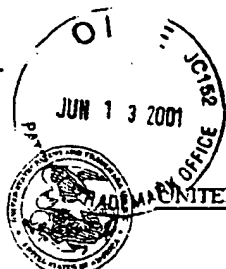
If there is any fee due in connection with the filing of this Request, please charge the fee to our Deposit Account No. 06-0916.

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW,  
GARRETT & DUNNER, L.L.P.

Dated: June 12, 2001

By:   
Phil M. Makrogiannis  
Reg. No. 47,766



UNITED STATES PATENT AND TRADEMARK OFFICE

 COMMISSIONER FOR PATENTS  
 UNITED STATES PATENT AND TRADEMARK OFFICE  
 WASHINGTON, D.C. 20231  
 www.uspto.gov

APPLICATION NUMBER	FILING DATE	GRP ART UNIT	FIL FEE REC'D	ATTY. DOCKET NO	DRAWINGS	TOT CLAIMS	IND CLAIMS
09/756,573	01/08/2001	1712	808	OC-573A1	2	21	4

CONFIRMATION NO. 6960

CORRECTED FILING RECEIPT



\*OC00000006042000\*

 PPG INDUSTRIES INC.  
 Intellectual Property Department  
 One PPG Place  
 Pittsburgh, PA 15272

Date Mailed: 05/04/2001

Receipt is acknowledged of this nonprovisional Patent Application. It will be considered in its order and you will be notified as to the results of the examination. Be sure to provide the U.S. APPLICATION NUMBER, FILING DATE, NAME OF APPLICANT, and TITLE OF INVENTION when inquiring about this application. Fees transmitted by check or draft are subject to collection. Please verify the accuracy of the data presented on this receipt. If an error is noted on this Filing Receipt, please write to the Office of Initial Patent Examination's Customer Service Center. Please provide a copy of this Filing Receipt with the changes noted thereon. If you received a "Notice to File Missing Parts" for this application, please submit any corrections to this Filing Receipt with your reply to the Notice. When the USPTO processes the reply to the Notice, the USPTO will generate another Filing Receipt incorporating the requested corrections (if appropriate).

## Applicant(s)

Susan E. Jonathan D. Zook, Santa Clarita, CA;  
~~Suzanne G.~~ DeMoss, Van Nuys, CA;  
 David W. Jordan, Northridge, CA;  
 Chandra B. Rao, Valencia, CA;  
 Hakam Singh, Bradbury, CA;  
 Ahmed Sharaby, Canyon Country, CA;

## Domestic Priority data as claimed by applicant

THIS APPLICATION IS A CIP OF 08/928,972 09/12/1997 PAT 6,172,179  
~~WHICH IS A CIP OF 09/318,500 05/25/1999 PAT 6,232,401~~  
 THIS APPLICATION 09/756,573  
 CLAIMS BENEFIT OF 60/215,548 06/30/2000  
 AND CLAIMS BENEFIT OF 60/182,396 02/14/2000

Which is a  
 CIP of 08/802,13  
 02/19/97 PAT  
 5,912,319.  
 This application is  
 also

which is a DIV  
 of 08/802,130  
 02/19/97 PAT  
 5,912,319.

## Foreign Applications

Projected Publication Date: To Be Determined - pending completion of Missing Parts

Non-Publication Request: No

Early Publication Request: No

### Title

### Sealants and potting formulations including mercapto-terminated polymers produced by the reaction of a polythiol and polyvinyl ether monomer

## Preliminary Class

528

**Data entry by : DADE JR, THEODORE**

**Team : OIPE**

Date: 05/04/2001

[illegible]

**LICENSE FOR FOREIGN FILING UNDER  
Title 35, United States Code, Section 184  
Title 37, Code of Federal Regulations, 5.11 & 5.15**

**GRANTED**

The applicant has been granted a license under 35 U.S.C. 184, if the phrase "IF REQUIRED, FOREIGN FILING LICENSE GRANTED" followed by a date appears on this form. Such licenses are issued in all applications where the conditions for issuance of a license have been met, regardless of whether or not a license may be required as set forth in 37 CFR 5.15. The scope and limitations of this license are set forth in 37 CFR 5.15(a) unless an earlier license has been issued under 37 CFR 5.15(b). The license is subject to revocation upon written notification. The date indicated is the effective date of the license, unless an earlier license of similar scope has been granted under 37 CFR 5.13 or 5.14.

This license is to be retained by the licensee and may be used at any time on or after the effective date thereof unless it is revoked. This license is automatically transferred to any related applications(s) filed under 37 CFR 1.53(d). This license is not retroactive.

The grant of a license does not in any way lessen the responsibility of a licensee for the security of the subject matter as imposed by any Government contract or the provisions of existing laws relating to espionage and the national security or the export of technical data. Licensees should apprise themselves of current regulations especially with respect to certain countries, of other agencies, particularly the Office of Defense Trade Controls, Department of State (with respect to Arms, Munitions and Implements of War (22 CFR 121-128)); the Office of Export Administration, Department of Commerce (15 CFR 370.10 (j)); the Office of Foreign Assets Control, Department of Treasury (31 CFR Parts 500+) and the Department of Energy.

**NOT GRANTED**

No license under 35 U.S.C. 184 has been granted at this time, if the phrase "IF REQUIRED, FOREIGN FILING LICENSE GRANTED" DOES NOT appear on this form. Applicant may still petition for a license under 37 CFR 5.12, if a license is desired before the expiration of 6 months from the filing date of the application. If 6 months has lapsed from the filing date of this application and the licensee has not received any indication of a secrecy order under 35 U.S.C. 181, the licensee may foreign file the application pursuant to 37 CFR 5.15 (b).

**PLEASE NOTE the following information about the Filing Receipt:**

- The articles such as "a," "an" and "the" are not included as the first words in the title of an application. They are considered to be unnecessary to the understanding of the title.
- The words "new," "improved," "improvements in" or "relating to" are not included as first words in the title of an application because a patent application, by nature, is a new idea or improvement.
- The title may be truncated if it consists of more than 500 characters (letters and spaces combined).
- The docket number allows a maximum of 25 characters.
- If your application was submitted under 37 CFR 1.10, your filing date should be the "date in" found on the Express Mail label. If there is a discrepancy, you should submit a request for a corrected Filing Receipt along with a copy of the Express Mail label showing the "date in."
- The title is recorded in sentence case.

Any corrections that may need to be done to your Filing Receipt should be directed to:

Assistant Commissioner for Patents  
Office of Initial Patent Examination  
Customer Service Center  
Washington, DC 20231

Filed on behalf of: Senior Party Jonathan D. Zook

By: Jerry D. Voight  
Wesley B. Derrick  
FINNEGAN, HENDERSON, FARABOW,  
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Telephone: 650.849.6600  
Facsimile: 650.849.6666

UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

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JONATHAN D. **ZOOK**, DAVID W. JORDAN, DEAN M. WILLARD,  
GEORGE JONES, AND MICHAEL COSMAN,  
Junior Party ("PBT")  
(U.S. Patents 6,525,168 and 6,723,827),

v.

JONATHAN D. **ZOOK**, SUSAN E. DEMOSS,  
DAVID W. JORDAN AND CHANDRA B. RAO  
Senior Party ("PRC")  
(U.S. Application 10/644,389).

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Patent Interference No. 105,555 (MPT)  
(Technology Center 1700)

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**DECLARATION OF PHILIPPE N. MAKROGIANNIS**  
**ZOOK EXHIBIT 1027**

**ZOOK EXHIBIT 1027**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

I, Philippe N. Makrogiannis, declare that:

1. I am a patent attorney registered to practice before the U.S. Patent and Trademark Office (USPTO). My registration number is 47,766.
2. I am also an active member of the State Bar of California.
3. I am currently employed as a patent attorney by Applied Biosystems, Inc., 850 Lincoln Centre Drive, Foster City, CA 94404.
4. Prior to my employment by Applied Biosystems, Inc., I was employed as a patent attorney from about November 1999 to November 2002 by the law firm of Finnegan, Henderson, Farabow, Garrett and Dunner (Finnegan) in their California office now located at 3300 Hillview Drive, Palo Alto, CA 94304.
5. During the time I was employed by Finnegan I was responsible for a number of patent matters for the firm client PRC-DeSoto International, Inc. (formerly Courtaulds Aerospace, Inc.), a wholly-owned subsidiary of PPG Industries.
6. Among the matters I worked on were U.S. Application Nos. 09/756,573, filed on January 8, 2001, and 10/302,177, filed on November 21, 2002. These applications are part of a chain of divisional, continuation, and continuation-in-part applications, the relationship of which is depicted on Zook Exhibit 1003. The various relevant applications and patents in the chain of applications referred to in Zook Exhibit 1003 are numbered 1 (for the application having the earliest filing date) to 6 (for the most recently filed application). For convenience, the various applications and patents will be referred

to herein by both the application or patent number and by the sequential numbers appearing in Zook Exhibit 1003.

7. Application No. 10/302,177 (No. 5) was filed with a preliminary amendment and request for interference on November 21, 2002. Zook Exhibit 1016. The application transmittal for the '177 application (No. 5) further directed the amendment of the specification to recite a priority claim that included Application No. 09/318,500 (No. 3).

8. I have been advised that Application No. 10/644,389 (No. 6) has been placed in interference with U.S. Patent Nos. 6,525,168 and 6,723,389 (respectively, the '168 and '187 patents).

9. I have also been advised that Application No. 10/644,389 (No. 6) includes an alternative priority chain that involves Application No. 09/318,500 (No. 3).

10. Specifically, the main chain of priority applications begins with Application No. 08/802,130 (No. 1), and runs through Application Nos. 08/928,972 (No. 2), 09/756,573 (No. 4), and 10/302,177 (No. 5) to the application currently in the interference, Application 10/644,389 (No. 6). The alternate chain of priority applications should also begin with Application No. 08/802,130 (No. 1) and run through Application Nos. 09/318,500 (No. 3), 09/756,573 (No. 4), and 10/302,177 (No. 5) to Application No. 10/644,389 (No. 6). See Zook Exhibit 1003 (in the exhibit, the correct chains of priority cases are connected by solid lines).

11. Application No. 09/756,573 (No. 4) (now U.S. Patent No. 6,509,418), the first application in the relevant chain of applications that could have claimed priority to

Application No. 09/318,500 (No. 3), properly recited both the alternative chain that includes Application No. 09/318,500 (No. 3) and the chain that does not. Specifically, in the alternative priority chain, as set forth in Application No. 09/756,573 (No. 4), that application is properly identified as a continuation-in-part of Application No. 09/318,500 (No. 3), which application is a divisional of Application No. 08/802,130 (No. 1). See Zook Exhibit 1014, first paragraph of Col 1.

12. During prosecution of the '573 application (No. 4), I became aware of an error in the Domestic Priority Data as claimed by Applicants in the Corrected Filing Receipt issued by the Patent Office and filed a Request for Corrected Filing Receipt on June 12, 2001. Exhibit 1026. This request correctly set forth, again, the relationship between the '573 application and the '500 application, which relationship is that set forth in the first paragraph of U.S. Patent No. 6,509,418, the patent which issued from the '573 application. See Zook Exhibit 1014, first paragraph of Col 1.

13. Application No. 10/302,177 (No. 5) was filed with a slight change in punctuation and wording to reflect the continuity from Application No. 09/756,573 (No. 4). See Zook Exhibits 1014 and 1007, paragraph 9, page 2. Because of the change in punctuation, along with minor changes in wording, one may be confused and not recognize from the alternative priority claim that Application No. 09/318,500 (No. 3) is a parent continuation-in-part of Application No. 09/756,573 (No. 4). Instead of recognizing the proper alternative priority, one may misinterpret the priority claim recited and read that Application No. 10/318,500 (No. 3) is a parent continuation-in-part of Application No. 10/302,177 (No. 5). Application No. 10/302,177 (No. 5) could not directly claim priority from Application No. 09/318,500 (No. 3) because the two applications were never

copending. In contrast, Application No. 09/318,500 (No. 3) and Application No. 09/756,573 (No. 4) were copending and the latter application could properly claim priority from the former application.

14. Subsequent to filing Application No. 10/302,177 (No. 5) on November 21, 2002, with the preliminary amendment and request for interference (Zook Exhibit 1016), I left the Finnegan Firm and had no further involvement with the prosecution of Application No. 10/302,177 (No. 5).

15. At the time of filing Application No. 10/302,177 (No. 5), I did not realize that the priority claim to the alternative chain of applications could be construed to incorrectly suggest that Application No. 10/302,177 (No. 5) claimed priority directly from Application No. 09/318,500 (No. 3) (the dash line on Zook Exhibit 1003). However, because these two applications were not copending, priority to Application 09/318,500 (No. 3) could only be obtained through a chain of continuation and continuation-in-part applications including Application No. 09/756,573 (No. 4).

16. The alternative priority claim involving Application No. 09/318,500 (No. 3) that could be improperly construed and misinterpreted was never intended to allow for improper interpretation; any shortcoming was inadvertent and unintentional. It was my intent at all times that there be a properly claimed alternative priority chain that included Application No. 09/318,500 (No. 3).

17. Moreover, at no time was I, nor to the best of my knowledge was anyone else affiliated with PRC, aware of any deficiency in the priority claims involving Application No. 09/318,500 (No. 3) until the defect in the alternative chain was discovered by

interference counsel after the interference was declared. Thus, the entire delay in making the correct priority chain involving Application 09/318,500 (No. 3) has remained unintentional at all times.

18. The correct alternative priority claim in both Application No. 10/302,177 (No. 5) and Application No. 10/644,389 (No. 6) should recite "Said Application No. 09/756,573 (No. 4) is also a continuation-in-part of Application No. 09/318,500 (No. 3), which is a divisional of Application No. 08/802,130 (No. 1)." Specifically, Application No. 09/756,573 (No. 4) is a continuation in-part of both Application No. 08/928,972 (No. 2) and Application No. 09/318,500 (No. 3).


19. That this deficiency was inadvertent, unintentional and made in good faith is evident in that it does not affect entitlement to benefit of the filing date of the earliest application in the chain, Application No. 08/802,130 (No. 1).

20. In signing this Declaration, I understand that the Declaration will be filed as evidence in a contested case before the Board of Patent Appeals and Interferences of the United States Patent and Trademark Office. I acknowledge that I may be subject to cross examination in the case and that cross examination will take place within the United States. If cross examination is required of me, I will appear for cross examination within the United States during the time allotted for cross examination.

21. The undersigned further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both,

under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing therefrom.

Date: 08 Nov 07

By:   
Philippe N. Makrogiannis

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re patent application of:  
Jonathan ZOOK et al.

Group Art Unit:

A Divisional Application of 08/802,130

Examiner:

Serial No. UNASSIGNED

Filed:

For: COMPOSITION AND METHOD  
FOR PRODUCING FUEL  
RESISTANT LIQUID  
POLYTHIOETHER POLYMERS  
WITH GOOD LOW  
TEMPERATURE FLEXIBILITY

CERTIFICATE OF MAILING

Assistant Commissioner for Patents  
Washington, D.C. 20231

I hereby certify that a Divisional Patent Application Under 37 CFR §1.53(b); Specification - 54 Pages; Transmittal Form 1082; Two (2) pages Formal Drawings; Copy of Declaration; and return postcard are being deposited with the United States Postal Service, postage prepaid, Express Mail No. EL147380746US on the date indicated below and addressed to: Assistant Commissioner for Patents, Washington, D.C. 20231.

May 25, 1999

(Date of Deposit)

DAVID J. MEYER

(Name of Person Mailing Paper or Fee)

David J. Meyer

(Signature)

LADOCS\2500368 1

**ZOOK EXHIBIT 1028**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

FORM PTO-1082  
ASSISTANT COMMISSIONER FOR PATENTS  
Washington, D.C. 20231

EXPRESS MAIL NO. EL147380746US

Case Docket No. 58179-5002

Date: May 25, 1999

Sir:

Transmitted herewith for filing is a Divisional patent application of 08/802,130 under 37 CFR §1.53(b) of:

Inventor(s): Jonathan Doherty ZOOK, Suzanna GIBSON, David Weldon JORDAN and Chandra B. RAO

For: COMPOSITION AND METHOD FOR PRODUCING FUEL RESISTANT LIQUID POLYTHIOETHER POLYMERS WITH GOOD LOW TEMPERATURE FLEXIBILITY

Enclosed are:

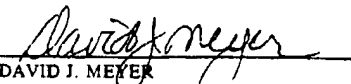
Two Sheet(s) of drawing ( ☒ formal ☐ informal ) +     extra copies

- ☐ An assignment of the invention to \_\_\_\_\_  
☐ Will follow.
- ☐ A certified copy of \_\_\_\_\_  
from which priority is claimed in the subject case pursuant to Rule 55(b) and 35 USC 119(a)-(d).  
☐ Will follow.
- ☐ Power of Attorney by Assignee and Exclusion of Inventor Under Rule 3.71.  
☐ A Verified Statement to Establish Small Entity Status under 37 CFR 1.9 and 37 CFR 1.27.  
☒ Copy of the Declaration  
☐ Recordation Form Cover Letter (Form PTO 1595)  
☐ Information Disclosure Statement (Form PTO 1449), with        attachments  
☐ Petition to Make Special under 37 CFR §1.102 and MPEP §708.02(VIII).

FOR:	NO. FILED	NO. EXTRA	SMALL ENTITY		OR	OTHER THAN SMALL ENTITY	
			RATE	FEE		RATE	FEE
BASIC FEE				\$ 380			\$ 760
TOTAL CLAIMS	24 - 20	4	x 9 =	\$	OR	x 18 =	\$ 72
INDEP CLAIMS	3		x 39 =	\$	OR	x 78 =	\$
MULTIPLE DEPENDENT CLAIMS PRESENTED			+ \$130			+ \$260	
TOTAL				\$			\$ 832

- ☐ Please charge my Deposit Account No. 10-0440 the amount of \$ \_\_\_\_\_. A duplicate copy of this sheet is enclosed.
- ☐ A check in the amount of \$ to cover the filing fee is enclosed.
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- ☐ Any patent application processing fees under 37 CFR 1.17.
- ☐ The issue fee set in 37 CFR 1.18 at or before mailing of the Notice of Allowance, pursuant to 37 CFR 1.311(b).
- ☐ Any filing fees under 37 CFR 1.16 for presentation of extra claims.

Respectfully submitted,

  
DAVID J. MEYER  
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Rev. 06/1/94

DECLARATION

☒ ORIGINAL  
☐ CONTINUATION  
☐ DIVISIONAL

As a below named inventor, I declare that the information given herein is true, that I believe that I am the original, first and sole inventor if only one name is listed at 1 below, or a joint inventor if plural inventors are named below at 1-4, of the invention entitled:  
COMPOSITION AND METHOD FOR PRODUCING FUEL RESISTANT LIQUID POLYTHIOETHER POLYMERS WITH GOOD LOW TEMPERATURE FLEXIBILITY

Which is described and claimed in:

- ☒ the attached specification or  
☐ the specification in application Serial No. \_\_\_\_\_ filed \_\_\_\_\_ ☐ as amended on \_\_\_\_\_ (if applicable)  
(for declaration not accompanying application)

and for which a patent is sought, and that my residence, post office address and citizenship are as stated below next to my name.  
I acknowledge my duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations §1.56(a).  
I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.  
I hereby claim foreign priority benefits under Title 35, United States Code, §119 (a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

PRIOR FOREIGN APPLICATION(S)			
COUNTRY	APPLICATION NUMBER	DATE OF FILING (day/month/year)	PRIORITY CLAIMED UNDER 35 UCS §119
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

I hereby claim the benefit under Title 35, United States Code, §119(a) of any United States provisional application(s) listed below:

APPLICATION SERIAL NO.	FILING DATE

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code §120, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

APPLICATION SERIAL NO.	FILING DATE	STATUS

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1	Name of Inventor	LAST NAME Zook	FIRST NAME Jonathan	MIDDLE NAME Ooherty	Residence: CITY	STATE or COUNTRY
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	Signature <i>Jonathan Ooherty Zook</i>				Date <i>January 31, 1997</i>	
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	Signature <i>Suzanna Gibson</i>				Date <i>January 31, 1997</i>	
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	Signature <i>David Weldon Jordan</i>				Date <i>January 31, 1997</i>	
4	Name of Inventor	LAST NAME Rao	FIRST NAME Chandra	MIDDLE NAME B.	Residence: CITY	STATE or COUNTRY
	Post Office Address 25716 North Player Dr. Valencia, Calif. 91355					CITIZENSHIP India
	Signature <i>Chandra Rao</i>				Date <i>January 31, 1997</i>	

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

(12) **United States Patent**  
Zook et al.

(10) Patent No.: **US 6,232,401 B1**  
(45) Date of Patent: **May 15, 2001**

(54) **COMPOSITION FOR PRODUCING FUEL RESISTANT LIQUID POLYTHIOETHER POLYMERS WITH GOOD LOW TEMPERATURE FLEXIBILITY**

(75) Inventors: **Jonathan Doherty Zook**, Santa Clarita;  
**Suzanna Gibson DeMoss**, Van Nuys;  
**David Weldon Jordan**, Reseda;  
**Chandra B. Rao**, Valencia, all of CA (US)

(73) Assignee: **PRC-DeSoto International, Inc.**, Glendale, CA (US)

(\*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days.

(21) Appl. No.: **09/318,500**

(22) Filed: **May 25, 1999**

**Related U.S. Application Data**

(62) Division of application No. 08/802,130, filed on Feb. 19, 1997, now Pat. No. 5,912,319.

(51) Int. Cl.<sup>7</sup> ..... **C08F 8/00**

(52) U.S. Cl. .... **525/191; 528/373; 528/374; 528/378; 568/29; 524/609**

(58) Field of Search ..... **528/373, 374, 528/378, 212; 568/29; 524/609; 525/212, 191**

(56) **References Cited**

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4,366,307	12/1982	Singh et al.	528/373
4,609,762	9/1986	Morris et al.	568/38
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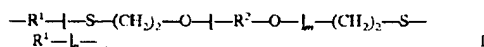
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Primary Examiner—Duc Truong

(74) Attorney, Agent, or Firm—Ann Marie Cannoni

(57) **ABSTRACT**

A polythioether includes a structure having the formula I



wherein

R<sup>1</sup> denotes a divalent C<sub>2-6</sub> n-alkyl, C<sub>3-6</sub> branched alkyl, C<sub>6-8</sub> cycloalkyl or C<sub>6-10</sub> alkylcycloalkyl group,  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}[(\text{---CH}_2\text{---})_r\text{---}]$ , or  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}[(\text{---CH}_2\text{---})_r\text{---}]$  in which at least one  $\text{---CH}_2\text{---}$  unit is substituted with a methyl group,

R<sup>2</sup> denotes methylene, a divalent C<sub>2-6</sub> n-alkyl, C<sub>2-6</sub> branched alkyl, C<sub>6-8</sub> cycloalkyl or C<sub>6-10</sub> alkylcycloalkyl group, or  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}[(\text{---CH}_2\text{---})_r\text{---}]$ ,

X denotes one selected from the group consisting of O, S and  $\text{---NR}^6$ ,

R<sup>6</sup> denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

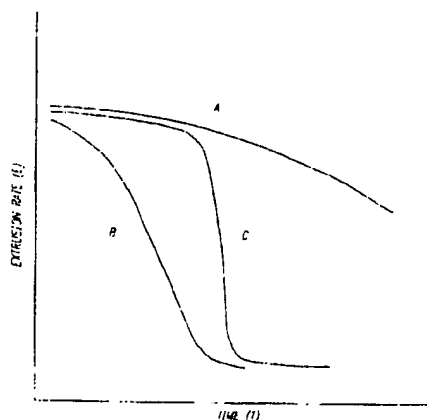
p is an integer from 2 to 6,

q is an integer from 0 to 5, and

r is an integer from 2 to 10.

The polythioether is a liquid at room temperature and pressure.

**11 Claims, 2 Drawing Sheets**



**ZOOK EXHIBIT 1029**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

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FIG. 1

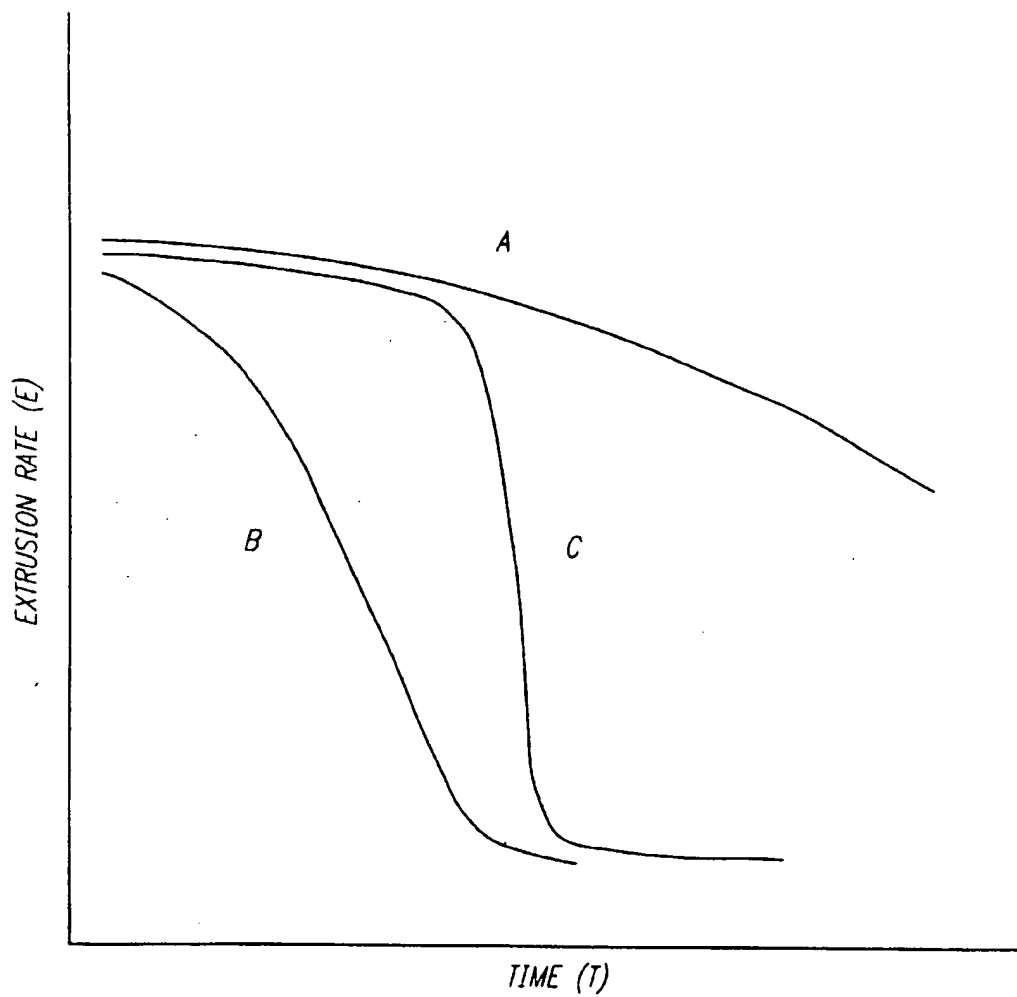
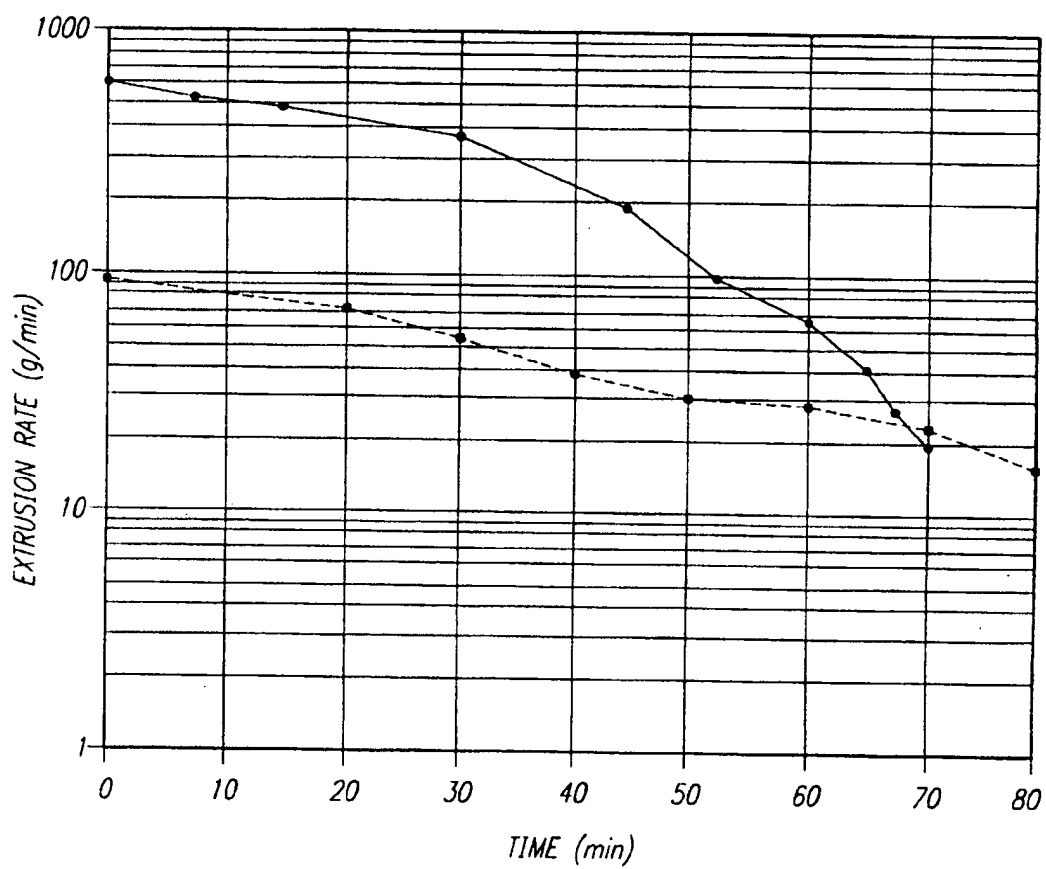


FIG. 2



# COMPOSITION FOR PRODUCING FUEL-RESISTANT LIQUID POLYTHIOETHER POLYMERS WITH GOOD LOW TEMPERATURE FLEXIBILITY

This application is a divisional of Ser. No. 08/802,130 filed Feb. 19, 1997 now U.S. Pat. No. 5,912,319.

## FIELD OF THE INVENTION

The present invention relates to liquid polythioether polymers that have good low temperature flexibility and fuel resistance when cured. The invention is also directed to methods for making the polymers by reacting polythiols with oxygenated dienes (divinyl ethers) which substantially eliminate malodorous condensed cyclic by-products.

## BACKGROUND OF THE INVENTION

Thiol-terminated sulfur-containing polymers are known to be well-suited for use in aerospace sealants due to their fuel resistant nature upon cross-linking. Among the commercially available polymeric materials which have sufficient sulfur content to exhibit this desirable property are the polysulfide polyformal polymers described, e.g., in U.S. Pat. No. 2,466,963, and the alkyl side chain containing polythioether polymers described, e.g., in U.S. Pat. No. 4,366,307 to Singh et al. Materials useful in this context also have the desirable properties of low temperature flexibility (low glass transition temperature  $T_g$ ) and liquidity at room temperature.

An additional desirable combination of properties for aerospace sealants which is much more difficult to obtain is the combination of long application time (i.e., the time during which the sealant remains usable) and short curing time (the time required to reach a predetermined strength). Singh et al., U.S. Pat. No. 4,366,307, disclose such materials. Singh et al. teach the acid-catalyzed condensation of hydroxyl-functional thioethers. The hydroxyl groups are in the  $\beta$ -position with respect to a sulfur atom for increased condensation reactivity. The Singh et al. patent also teaches the use of hydroxyl-functional thioethers with pendent methyl groups to afford polymers having good flexibility and liquidity. However, the disclosed condensation reaction has a maximum yield of about 75% of the desired condensation product. Furthermore, the acid-catalyzed reaction of  $\beta$ -hydroxysulfide monomers yields significant quantities (typically not less than about 25%) of an aqueous solution of thermally stable and highly malodorous cyclic byproducts, such as 1-thia-4-oxa-cyclohexane. As a result, the commercial viability of the disclosed polymers is limited.

Another desirable feature in polymers suitable for use in aerospace sealants is high temperature resistance. Inclusion of covalently bonded sulfur atoms in organic polymers has been shown to enhance high temperature performance. However, in the polysulfide polyformal polymers disclosed in U.S. Pat. No. 2,466,963, the multiple  $-S-S-$  linkages in the polymer backbones result in compromised thermal resistance. In the polymers of Singh et al., U.S. Pat. No. 4,366,307, enhanced thermal stability is achieved through replacement of polysulfide linkages with polythioether ( $-S-$ ) linkages. In practice, however, the disclosed materials also have compromised thermal resistance due to traces of the residual acid condensation catalyst.

Morris et al., U.S. Pat. No. 4,609,762, describes reacting dithiols with secondary or tertiary alcohols to afford liquid polythioethers having no oxygen in the polymeric backbone. Cured polymeric materials formed from these polymers

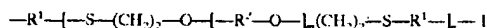
have the disadvantage, however, of reduced fuel resistance due to the large number of pendent methyl groups that are present. In addition, residual catalyst from the disclosed process generates undesirable aqueous acidic waste.

Cameron, U.S. Pat. No. 5,225,472, discloses production of polythioether polymers by the acid-catalyzed condensation of dithiols with active carbonyl compounds such as  $HCOOH$ . Again, this process generates undesirable aqueous acidic waste.

The addition polymerization of aliphatic dithiols with diene monomers has been described in the literature. See, e.g., Klemm, E. et al., *J. Macromol. Sci.—Chem.*, A28(9), pp. 875–883 (1991); Nuyken, O. et al., *Makromol. Chem., Rapid Commun.* 11, 365–373 (1990). However, neither Klemm et al. nor Nuyken suggest selection of particular starting materials, specifically divinyl ethers and dithiols, such that a polymer is formed that is liquid at room temperature and, upon curing, has excellent low-temperature flexibility (low  $T_g$ ) and high resistance to fuels, i.e., hydrocarbon fluids. Nor do Klemm et al. suggest production of a polymer that in addition is curable at room or lower temperatures. Moreover, the reactions disclosed by Klemm et al. also generate undesirable cyclic byproducts.

## SUMMARY OF THE PREFERRED EMBODIMENTS

In accordance with one aspect of the present invention, there is provided a polythioether having the formula I



wherein

$R^1$  denotes a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $-[(CH_2)_p-X]_q-$  or  $-[(CH_2)_p-X]_q-$  in which at least one  $-CH_2-$  unit is substituted with a methyl group,

$R^2$  denotes methylene, a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group, or  $-[(CH_2)_p-X]_q-$  in which at least one  $-CH_2-$  unit is substituted with a methyl group,

X denotes one selected from the group consisting of O, S and  $-NR^6-$ ,

$R^6$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10,

the polythioether being a liquid at room temperature and pressure.

Preferably the polythioether has a number average molecular weight between about 500 and about 20,000.

In a first preferred embodiment, the polythioether has the formula II



wherein

A denotes a structure having the formula I,

y is 0 or 1,

$R^3$  denotes a single bond when  $y=0$

and  $-S-(CH_2)_2-[-O-R^2-]_m-O-$  when  $y=1$ ,

$R^4$  denotes  $-SH$  or  $-S-(CH_2)_2-O-R^5$  when  $y=0$

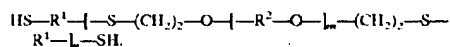
and  $-CH_2=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,

$R^5$  denotes  $C_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $-OH$  or  $-NHR^7$  group, and

$R^7$  denotes H or a  $C_{1-6}$  n-alkyl group.

Polythioethers in which  $R^4$  is  $-\text{SH}$  are "uncapped," that is, include unreacted terminal thiol groups. Polythioethers according to the invention also include "capped" polythioethers, that is, polythioethers including terminal groups other than unreacted thiol groups. These terminal groups can be groups such as  $-\text{OH}$  or  $-\text{NH}_2$ , or groups such as alkyl or terminal ethylenically unsaturated groups.

In a more particular preferred embodiment,  $y=0$  in formula II and  $R^4$  denotes  $-\text{SH}$ . That is, the polythioether is an uncapped polythioether having the structure

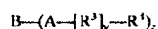


In another more particular preferred embodiment, the inventive polythioether is a capped polythioether in which  $y=0$  in formula II and  $R^4$  denotes  $-\text{S}-(\text{CH}_2)_2-\text{O}-\text{R}^5$ . Particularly preferably,  $R^5$  is an unsubstituted or substituted n-alkyl group such as ethyl, 4-hydroxybutyl or 3-aminopropyl.

In still another particular preferred embodiment,  $y=1$  in formula II and  $R^4$  denotes  $-\text{CH}=\text{CH}_2$ . That is, the polythioether is an uncapped polythioether having terminal vinyl groups.

In yet another more particular preferred embodiment, the inventive polythioether is a capped polythioether in which  $y=1$  in formula II and  $R^4$  denotes  $-(\text{CH}_2)_2-\text{S}-\text{R}^5$ .

In a second preferred embodiment, the polythioether has the formula III



wherein

A denotes a structure having the formula I,

y is 0 or 1,

$R^3$  denotes a single bond when  $y=0$

and  $-\text{S}-(\text{CH}_2)_2-[\text{O}-\text{R}^2-]_m-\text{O}-$  when  $y=1$ ,

$R^4$  denotes  $-\text{SH}$  or  $-\text{S}-(\text{CH}_2)_2-\text{O}-\text{R}^5$  when  $y=0$

and  $-\text{CH}_2=\text{CH}_2$  or  $-(\text{CH}_2)_2-\text{S}-\text{R}^5$  when  $y=1$ ,

$R^5$  denotes  $\text{C}_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $-\text{OH}$  or  $-\text{NHR}^7$  group,

$R^7$  denotes H or a  $\text{C}_{1-6}$  n-alkyl group.

z is an integer from 3 to 6, and

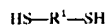
B denotes a z-valent residue of a polyfunctionalizing agent.

That is, the polyfunctionalized embodiments include three or more structures of the formula I bound to the residue of an appropriate polyfunctionalizing agent.

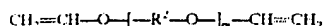
In a more specific embodiment, z is 3, and the polyfunctionalizing agent thus is a trifunctionalizing agent. In another more specific embodiment, the average functionality of the polythioether ranges between about 2.05 and about 3.00.

In accordance with another aspect of the present invention, there are provided methods of producing the foregoing polythioethers.

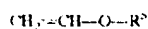
According to a first preferred embodiment, a polythioether of the invention is produced by reacting (n+1) equivalents (e.g., moles) of a compound having the formula IV



or a mixture of at least two different compounds having the formula IV, with (n) equivalents of a compound having the formula V



or a mixture of at least two different compounds having the formula V, and optionally about 0.05 to about 2 equivalents of a compound having the formula VI



or a mixture of two different compounds having the formula VI, in the presence of a catalyst. The catalyst is selected from the group consisting of free-radical catalysts, ionic catalysts and ultraviolet light. Preferably the catalyst is a free-radical catalyst such as an azo compound.

According to a second preferred embodiment, a polythioether of the invention is produced by reacting (n) equivalents of a compound having the formula IV, or a mixture of at least two different compounds having the formula IV, with (n+1) equivalents of a compound having the formula V, or a mixture of at least two different compounds having the formula V, optionally together with 0.05 to about 2 equivalents of a compound having the formula VII



or a mixture of two different compounds having the formula VII, in the presence of a catalyst as described above.

Analogous methods for producing polyfunctional polythioethers using the foregoing reactants together with appropriate polyfunctionalizing agents are also provided.

Polythioethers produced by the foregoing methods are also provided.

In accordance with yet another aspect of the present invention, there is provided a polymerizable composition comprising (i) about 30 to about 90 wt % of at least one polythioether as defined herein, said at least one polythioether having a glass transition temperature not greater than  $-55^\circ\text{C}$ ., (ii) a curing agent in an amount from about 90 to about 150% of stoichiometric based on the amount of said at least one polythioether, and (iii) about 5 to about 60 wt % of a filler, with all wt % being based on the total weight of non-volatile components of the composition. The inventive composition is curable at a temperature of  $0^\circ\text{C}$ . or higher, preferably at a temperature of  $-20^\circ\text{C}$ . or higher.

In accordance with an additional aspect of the present invention, there is provided a polymerizable composition comprising (i) about 30 to about 90 wt % of at least one polythioether as defined herein, said at least one polythioether having a glass transition temperature not greater than  $-50^\circ\text{C}$ ., (ii) a curing agent in an amount from about 90 to about 150% of stoichiometric based on the amount of said at least one polythioether, (iii) a plasticizer in an amount from about 1 to about 40 wt %, and (iv) a filler in an amount from about 5 to about 60 wt %, with all wt % being based on the total weight of non-volatile components of the composition. The composition is curable at a temperature of  $0^\circ\text{C}$ . or higher, preferably at a temperature of  $-20^\circ\text{C}$ . or higher.

Cured polymeric materials prepared by polymerization of the foregoing compositions are also provided.

Other objects, features and advantages of the present invention will become apparent to those skilled in the art from the following detailed description. It is to be understood, however, that the detailed description and specific examples, while indicating preferred embodiments of the present invention, are given by way of illustration and not limitation. Many changes and modifications within the scope of the present invention may be made without departing from the spirit thereof, and the invention includes all such modifications.

#### BRIEF DESCRIPTION OF THE DRAWINGS

The invention may be more readily understood by referring to the accompanying drawings in which

FIG. 1 depicts linear graphs of extrusion rate (E) versus time (T) for sealant compositions of the invention in com-

parison to extrusion rate curves for known types of sealant composition, and

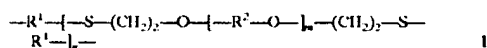
FIG. 2 is a semi-log graph of the extrusion rate curve of a polythioether of the invention (♦) and a prior art polysulfide (■).

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

It has surprisingly been discovered that the combination of certain polythiols with oxygenated dienes according to the present invention results in polythioether polymers that are liquids at room temperature and pressure and that have desirable physical and rheological properties, and that furthermore are substantially free of malodorous cyclic by-products. The inventive materials are also substantially free of deleterious catalyst residues, and hence have superior thermal resistance properties.

According to the present invention, polythioethers are provided that are liquid at room temperature and pressure and have excellent low temperature flexibility (low  $T_g$ ) and fuel resistance. As used herein, the term "room temperature and pressure" denotes approximately 77° F. (25° C.), and 1 atmosphere.

In their most general aspect, the inventive polythioethers include a structure having the formula I



wherein

$\text{R}^1$  denotes a divalent  $\text{C}_{2-6}$  n-alkyl,  $\text{C}_{3-6}$  branched alkyl,  $\text{C}_{6-8}$  cycloalkyl or  $\text{C}_{6-10}$  alkylcycloalkyl group,  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}[(\text{—CH}_2\text{—})_r\text{—}]$ , or  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}[(\text{—CH}_2\text{—})_r\text{—}]$  in which at least one  $\text{—CH}_2\text{—}$  unit is substituted with a methyl group,

$\text{R}^2$  denotes methylene, a divalent  $\text{C}_{2-6}$  n-alkyl,  $\text{C}_{2-6}$  branched alkyl,  $\text{C}_{6-8}$  cycloalkyl or  $\text{C}_{6-10}$  alkylcycloalkyl group, or  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}[(\text{—CH}_2\text{—})_r\text{—}]$ ,

X denotes one selected from the group consisting of O, S and  $\text{—NR}^3\text{—}$ ,

$\text{R}^3$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10.

Preferably, a polythioether polymer according to the invention has a glass transition temperature  $T_g$  that is not higher than  $-50^\circ\text{C}$ . More preferably, the  $T_g$  of the inventive polymer is not higher than  $-55^\circ\text{C}$ . Very preferably, the  $T_g$  of the inventive polymer is not higher than  $-60^\circ\text{C}$ . Low  $T_g$  is indicative of good low temperature flexibility, which can be determined by known methods, for example, by the methods described in AMS (Aerospace Material Specification) 3267 §4.5.4.7, MIL-S (Military Specification)-8802E §3.3.12 and MIL-S-29574, and by methods similar to those described in ASTM (American Society for Testing and Materials) D522-88.

The polythioethers of the invention exhibit very desirable fuel resistance characteristics when cured. One measure of the fuel resistance of the inventive polymers is their percent volume swell after prolonged exposure to a hydrocarbon fuel, which can be quantitatively determined using methods similar to those described in ASTM D792 or AMS 3269. Thus, in a preferred embodiment, the inventive polymers have, when cured, a percent volume swell not greater than 25% after immersion for one week at  $140^\circ\text{F}$ . ( $60^\circ\text{C}$ .) and

ambient pressure in jet reference fluid (JRF) type 1. Very preferably, the percent volume swell of the cured polymers is not greater than 20%.

JRF type 1, as employed herein for determination of fuel resistance, has the following composition (see AMS 2629, issued Jul. 1, 1989), section 3.1.1 et seq., available from SAE (Society of Automotive Engineers, Warrendale, Pa.):

Toluene	28 ± 1% by volume
Cyclohexane (technical)	34 ± 1% by volume
Isooctane	38 ± 1% by volume
Tertiary dibutyl disulfide (doctor sweet)	1 ± 0.005% by volume
Tertiary butyl mercaptan	0.015% ± 0.0015 by weight of the other four components

Desirably, the inventive polythioethers have number average molecular weights ranging from about 500 to 20,000, preferably about 1,000 to 10,000, very preferably about 2,000 to 5,000.

Liquid polythioether polymers within the scope of the present invention can be difunctional, that is, linear polymers having two end groups, or polyfunctional, that is, branched polymers having three or more end groups. Depending on the relative amounts of dithiol(s) and divinyl ether(s) used to prepare the polymers, the polymers can have terminal thiol groups ( $\text{—SH}$ ) or terminal vinyl groups ( $\text{—CH=CH}_2$ ). Furthermore, the polymers can be uncapped, that is, include thiol or vinyl terminal groups that are not further reacted, or capped, that is, include thiol or vinyl groups that are further reacted with other compounds. Capping the polythioethers of the invention enables introduction of additional terminal functionalities, for example, hydroxyl or amine groups, to the inventive polymers, or in the alternative, introduction of end groups that resist further reaction, such as terminal alkyl groups.

A first preferred embodiment of the inventive polythioethers has the formula II



wherein

A denotes a structure having the formula I,

y is 0 or 1,

$\text{R}^3$  denotes a single bond when  $y=0$

and  $\text{—S—}(\text{CH}_2)_2\text{—}[(\text{—O—R}^2)]_m\text{—O—}$  when  $y=1$ ,

$\text{R}^4$  denotes  $\text{—SH}$  or  $\text{—S—}(\text{CH}_2)_2\text{—O—R}^5$  when  $y=0$

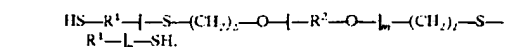
and  $\text{—CH}_2\text{=CH}_2$  or  $\text{—}(\text{CH}_2)_2\text{—S—R}^5$  when  $y=1$ ,

$\text{R}^5$  denotes  $\text{C}_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $\text{—OH}$  or  $\text{—NHR}^7$  group, and

$\text{R}^7$  denotes H or a  $\text{C}_{1-6}$  n-alkyl group.

Thus, polythioethers of the formula II are linear, difunctional polymers which can be uncapped or capped. When  $y=0$ , the polymer includes terminal thiol groups or capped derivatives thereof. When  $y=1$ , the polymer includes terminal vinyl groups or capped derivatives thereof.

According to one preferred embodiment, the inventive polythioether is a difunctional thiol-terminated (uncapped) polythioether. That is, in formula II,  $y=0$  and  $\text{R}^4$  is  $\text{—SH}$ . Thus, the polythioether has the following structure:



The foregoing polymers are produced, for example, by reacting a divinyl ether or mixture thereof with an excess of a dithiol or mixture thereof, as discussed in detail below.

In a more particular preferred embodiment of the foregoing polythioether, when  $m=1$  and  $\text{R}^2=n\text{-butyl}$  in formula II,

$R^1$  is not ethyl or n-propyl. Also preferably, when  $m=1$ ,  $p=2$ ,  $q=2$ ,  $r=2$  and  $R^2$ =ethyl, X is not O.

According to another preferred embodiment, the inventive polythioether is a capped polymer in which the foregoing terminal —SH groups are replaced by  $-S-(CH_2)_2-O-R^5$ . Such caps are produced by reaction of the terminal thiol group with a monovinyl ether, for example by including in the reaction mixture a capping agent or mixture thereof, as discussed in detail below.

In the foregoing,  $R^5$  denotes an unsubstituted or substituted alkyl group, preferably a  $C_{1-6}$  n-alkyl group which is unsubstituted or substituted with at least one —OH or —NHR<sup>7</sup> group, with  $R^7$  denoting H or  $C_{1-6}$  n-alkyl. Exemplary useful  $R^5$  groups include alkyl groups, such as ethyl, propyl and butyl; hydroxyl-substituted groups such as 4-hydroxybutyl; amine-substituted groups such as 3-aminopropyl; etc.

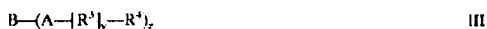
Polythioethers according to the invention also include difunctional vinyl-terminated (uncapped) polythioethers. That is, in formula II,  $y=1$  and  $R^4$  is  $-CH=CH_2$ . These polymers are produced, for example, by reacting a dithiol or mixture thereof with an excess of a divinyl ether or mixture thereof, as discussed in detail below. Analogous capped polythioethers include terminal  $-(CH_2)_2-S-R^5$ .

The foregoing polythioethers are linear polymers having a functionality of 2 (considering alkyl and other non-reactive caps within this total). Polythioethers having higher functionality are also within the scope of the present invention. Such polymers are prepared, as discussed in detail below, by using a polyfunctionalizing agent. The term "polyfunctionalizing agent" as employed herein denotes a compound having more than two moieties that are reactive with terminal —SH and/or  $-CH=CH_2$  groups. The polyfunctionalizing agent preferably includes from 3 to 6 such moieties, and thus is denoted a "z-valent" polyfunctionalizing agent, where z is the number (preferably from 3 to 6) of such moieties included in the agent, and hence the number of separate branches which the polyfunctional polythioether comprises. The polyfunctionalizing agent can be represented by the formula



where  $R^6$  denotes a moiety that is reactive with terminal —SH or  $-CH=CH_2$  and can be the same or different, and B is the z-valent residue of the polyfunctionalizing agent, i.e., the portion of the agent other than the reactive moieties  $R^6$ .

Polyfunctional polythioethers according to the present invention thus preferably have the formula III



wherein

A denotes a structure having the formula I,

y is 0 or 1,

$R^3$  denotes a single bond when  $y=0$

and  $-S-(CH_2)_2-[O-R^2]_m-O-$  when  $y=1$ ,

$R^4$  denotes —SH or  $-S-(CH_2)_2-O-R^5$  when  $y=0$

and  $-CH_2=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,

$R^5$  denotes  $C_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one —OH or —NHR<sup>7</sup> group,

$R^7$  denotes H or a  $C_{1-6}$  n-alkyl group,

z is an integer from 3 to 6, and

B denotes a z-valent residue of a polyfunctionalizing agent.

As with the preceding difunctional embodiments, the foregoing polyfunctional polythioethers of the present invention can include terminal —SH or  $-CH=CH_2$

groups, or can be capped and thus include terminal  $-S-(CH_2)_2-O-R^5$  or  $-(CH_2)_2-S-R^5$  groups. Partially capped polyfunctional polymers, i.e., polymers in which some but not all of the branches are capped, are also within the scope of the present invention.

Specific polyfunctionalizing agents include trifunctionalizing agents, that is, compounds with  $z=3$ . Preferred trifunctionalizing agents include triallylcyanurate (TAC), which is reactive with compounds of the formula II ( $R^6$ =allyl), and 1,2,3-propanetriethiol, which is reactive with compounds of the formula III ( $R^6$ =—SH). Agents having mixed functionality, i.e., agents that include moieties (typically separate moieties) that react with both thiol and vinyl groups, can also be employed.

Other useful polyfunctionalizing agents include trimethylolpropane trivinyl ether, and the polythiols described in U.S. Pat. No. 4,366,307, U.S. Pat. No. 4,609,762 and U.S. Pat. No. 5,225,472, the disclosures of each of which are incorporated in their entireties herein by reference.

Polyfunctionalizing agents having more than three reactive moieties (i.e.,  $z>3$ ) afford "star" polythioethers and hyperbranched polythioethers. For example, two equivalents of TAC can be reacted with one mole of a dithiol to afford a material having an average functionality of 4. This material can then be reacted with a divinyl ether and a dithiol to yield a polymer, which can in turn be mixed with a trifunctionalizing agent to afford a polymer blend having an average functionality between 3 and 4.

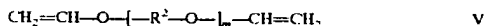
Polythioethers as described above have a wide range of average functionality. For example, trifunctionalizing agents afford average functionalities from about 2.05 to 3.0, preferably about 2.1 to 2.6. Wider ranges of average functionality can be achieved by using quaternary or higher polyfunctionalizing agents. Functionality will also be affected by factors such as stoichiometry, as is known to those skilled in the art.

Methods of making the foregoing polyfunctional polythioethers are discussed in detail below.

Polythioethers within the scope of the present invention are prepared by a number of methods. According to a first preferred method,  $(n+1)$  equivalents of a compound having the formula IV



or a mixture of at least two different compounds having the formula IV, are reacted with n equivalents of a compound having the formula V



or a mixture of at least two different compounds having the formula V, in the presence of a catalyst. In formulas IV and V above,  $R^1$ ,  $R^2$  and all indices are defined as in formula I. This method affords an uncapped, thiol-terminated difunctional polythioether.

The compounds of formula IV are dithiol compounds. Preferred dithiols include those compounds in which  $R^1$  is a divalent  $C_{2-6}$  n-alkyl group, i.e., 1,2-ethanedithiol, 1,3-propanedithiol, 1,4-butanedithiol, 1,5-pentanedithiol or 1,6-hexanedithiol.

Additional preferred dithiols include those compounds in which  $R^1$  is a divalent  $C_{3-6}$  branched alkyl group, having one or more pendent groups which can be, for example, methyl or ethyl groups. Preferred compounds having branched alkyl  $R^1$  include 1,2-propanedithiol, 1,3-butanedithiol, 2,3-butanedithiol, 1,3-pentanedithiol, 1,3-dithio-3-methylbutane and 2,3-butanedithiol. Other useful dithiols

include those in which  $R^1$  is a divalent  $C_{6-10}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group, for example, dipentenedimercaptan and ethylcyclohexyldithiol (ECHDT).

Further preferred dithiols include one or more heteroatom substituents in the carbon backbone, that is, dithiols in which  $X$  is a heteroatom such as O, S or another bivalent heteroatom radical; a secondary or tertiary amine group, i.e.,  $-NR^a-$ , where  $R^a$  is hydrogen or methyl; or another substituted trivalent heteroatom. In a preferred embodiment,  $X$  is O or S, and thus  $R^1$  is  $-[(CH_2)_p-O]_q-(CH_2)_r-$  or  $-[(CH_2)_p-S]_q-(CH_2)_r-$ . Preferably, the indices  $p$  and  $r$  are equal, and very preferably both have the value of 2. Particularly preferred exemplary dithiols of this type include dimercaptodiethylsulfide (DMDS) ( $p, r=2, q=1, X=S$ ); dimercaptodioxaoctane (DMDO) ( $p, q, r=2, X=O$ ); and 1,5-dithio-3-oxapentane. It is also possible to employ dithiols that include both heteroatom substituents in the carbon backbone and pendent alkyl, in particular methyl, groups. Such compounds include methyl-substituted DMDS, such as  $HS-CH_2CH(CH_3)-S-CH_2CH_2-SH$ ,  $HS-CH(CH_3)CH_2-S-CH_2CH_2-SH$  and dimethyl substituted DMDS such as  $HS-CH_2CH(CH_3)-S-CH(CH_3)CH_2-SH$  and  $HS-CH(CH_3)CH_2-S-CH_2CH(CH_3)-SH$ .

Two or more different dithiols of formula IV can also be employed if desired in preparing polythioethers according to the invention.

The compounds of formula V are divinyl ethers. Divinyl ether itself ( $m=0$ ) can be used. Preferred divinyl ethers include those compounds having at least one alkoxy group, more preferably from 1 to 4 alkoxy groups (i.e., those compounds in which  $m$  is an integer from 1 to 4). Very preferably,  $m$  is an integer from 2 to 4. It is also possible to employ commercially available divinyl ether mixtures in producing polythioethers according to the invention. Such mixtures are characterized by a non-integral average value for the number of alkoxy units per molecule. Thus,  $m$  in formula V can also take on non-integral, rational values between 0 and 10, preferably between 1 and 10, very preferably between 1 and 4, particularly between 2 and 4.

Exemplary divinyl ethers include those compounds in which  $R^2$  is  $C_{2-6}$  n-alkyl or  $C_{2-6}$  branched alkyl. Preferred divinyl ethers of this type include ethylene glycol divinyl ether (EG-DVE) ( $R^2$ =ethyl,  $m=1$ ); butanediol divinyl ether (BD-DVE) ( $R^2$ =butyl,  $m=1$ ); hexanediol divinyl ether (HD-DVE) ( $R^2$ =hexyl,  $m=1$ ); diethylene glycol divinyl ether (DEG-DVE) ( $R^2$ =ethyl,  $m=2$ ); triethylene glycol divinyl ether ( $R^2$ =ethyl,  $m=3$ ); and tetraethylene glycol divinyl ether ( $R^2$ =ethyl,  $m=4$ ). Useful divinyl ether blends include "PLURIOL®" type blends such as PLURIOL® E-200 divinyl ether (commercially available from BASF), for which  $R^2$ =ethyl and  $m=3.8$ , as well as "DPE" polymeric blends such as DPE-2 and DPE-3 (commercially available from International Specialty Products, Wayne, N.J.). Of these, DEG-DVE and PLURIOL® E-200 are particularly preferred.

Useful divinyl ethers in which  $R^2$  is  $C_{2-6}$  branched alkyl can be prepared by reacting a polyhydroxy compound with acetylene. Exemplary compounds of this type include compounds in which  $R^2$  is an alkyl-substituted methylene group such as  $-CH(CH_3)-$  or  $-CH_2CH(CH_3)-$ .

Other useful divinyl ethers include compounds in which  $R^2$  is polytetrahydrofuryl (poly-THF) or polyoxyalkylene, preferably having an average of about 3 monomer units.

Two or more compounds of the formula V can be used in the foregoing method. Thus in preferred embodiments of the invention, two compounds of formula IV and one com-

pounds of formula V, one compound of formula IV and two compounds of formula V, two compounds of formula IV and of formula V, and more than two compounds of one or both formulas, can be used to produce a variety of polythioethers according to the invention, and all such combinations of compounds are contemplated as being within the scope of the invention.

Although, as indicated above, compounds of the formulas IV and V which have pendent alkyl groups, for example pendent methyl groups, are useful according to the invention, it has surprisingly been discovered that compounds of the formulas IV and V which are free of pendent methyl or other alkyl groups also afford polythioethers that are liquid at room temperature and pressure.

The reaction between the compounds of formulas IV and V is preferably catalyzed by a free radical catalyst. Preferred free radical catalysts include azo compounds, for example azobisisnitrile compounds such as azo(bis)isobutyronitrile (AIBN); organic peroxides such as benzoyl peroxide and *t*-butyl peroxide; and inorganic peroxides such as hydrogen peroxide. The reaction can also be effected by irradiation with ultraviolet light either with or without a cationic photoinitiating moiety. Ionic catalysis methods, using either inorganic or organic bases, e.g., triethylamine, also yield materials useful in the context of this invention.

Capped analogs to the foregoing polythioethers can be prepared by reacting  $(n+1)$  equivalents of a compound having the formula IV or a mixture of at least two different compounds having the formula IV,  $(n)$  equivalents of a compound having the formula V or a mixture of at least two different compounds having the formula V, and about 0.05 to about 2 equivalents of a compound having the formula VI



or a mixture of two different compounds having the formula VI, in the presence of an appropriate catalyst.

Compounds of the formula VI are monovinyl ethers, which react with terminal thiol groups to cap the polythioether polymer. Preferred monovinyl ethers of the formula VI include amino- and hydroxyalkyl vinyl ethers, such as 3-aminopropyl vinyl ether and 4-hydroxybutyl vinyl ether (butanediol monovinyl ether), as well as unsubstituted alkyl vinyl ethers such as ethyl vinyl ether. Use of 2 equivalents of compounds of the formula VI affords fully capped polymers, while use of lesser amounts results in partially capped polymers.

According to another preferred method,  $(n)$  equivalents of a compound having the formula IV, or a mixture of at least two different compounds having the formula IV, are reacted with  $(n+1)$  equivalents of a compound having the formula V, or a mixture of at least two different compounds having the formula V, again in the presence of an appropriate catalyst. This method affords an uncapped, vinyl-terminated difunctional polythioether.

Capped analogs to the foregoing vinyl-terminated polythioethers can be prepared by reacting  $(n+1)$  equivalents of a compound having the formula V or a mixture of at least two different compounds having the formula V,  $(n)$  equivalents of a compound having the formula IV or a mixture of at least two different compounds having the formula IV, and about 0.05 to about 2 equivalents of a compound having the formula VII



or a mixture of two different compounds having the formula VII, in the presence of an appropriate catalyst.

Compounds of the formula VII are monothiois, which can be unsubstituted or substituted with, e.g., hydroxyl or amino groups. Exemplary capping compounds of the formula VII include mercaptoalcohols such as 3-mercaptoopropanol, and mercaptoamines such as 4-mercaptobutylamine.

Polyfunctional analogs of the foregoing difunctional polythioethers are similarly prepared by combining one or more compounds of formula IV and one or more compounds of formula V, in appropriate amounts, with a polyfunctionalizing agent as described above, and reacting the mixture. Thus, according to one method for making polyfunctional polythioethers of the present invention, (n+1) equivalents of a compound or compounds having the formula IV, (n) equivalents of a compound or compounds having the formula V, and a z-valent polyfunctionalizing agent, are combined to form a reaction mixture. The mixture is then reacted in the presence of a suitable catalyst as described above to afford thiol-terminated polyfunctional polythioethers. Capped analogs of the foregoing polythioethers are prepared by inclusion in the starting reaction mixture of about 0.05 to about (z) equivalents one or more appropriate capping compounds VI. Use of (z) equivalents affords fully capped polyfunctional polymers, while use of lesser amounts again yields partially capped polymers.

Similarly, (n) equivalents of a compound or compounds having the formula IV, (n+1) equivalents of a compound or compounds having the formula V, and a z-valent polyfunctionalizing agent, are combined to form a reaction mixture and reacted as above to afford vinyl-terminated polyfunctional polythioethers. Capped analogs of the foregoing polythioethers are prepared by inclusion in the starting reaction mixture of one or more appropriate capping compounds VII.

The inventive polythioethers preferably are prepared by combining at least one compound of formula IV and at least one compound of formula V, optionally together with one or capping compounds VI and/or VII as appropriate, and/or a polyfunctionalizing agent, followed by addition of an appropriate catalyst, and carrying out the reaction at a temperature from about 30 to about 120° C. for a time from about 2 to about 24 hours. Very preferably the reaction is carried out at a temperature from about 70 to about 90° C. for a time from about 2 to about 6 hours.

Since the inventive reaction is an addition reaction, rather than a condensation reaction, the reaction typically proceeds substantially to completion, i.e., the inventive polythioethers are produced in yields of approximately 100%. No or substantially no undesirable by-products are produced. In particular, the reaction does not produce appreciable amounts of malodorous cyclic by-products such as are characteristic of known methods for producing polythioethers. Moreover, the polythioethers prepared according to the invention are substantially free of residual catalyst. As a result, no free catalyst is available to further react with the polythioether, in particular in the presence of water at room temperature, to degrade the polymer and produce malodorous cyclic compounds. Thus, the inventive polythioethers are characterized both by thermal stability and by low odor.

Polythioethers according to the invention are useful in applications such as coatings and sealant compositions, and preferably are formulated as polymerizable sealant compositions in applications where low temperature flexibility and fuel resistance are important. Such sealant compositions are useful, e.g., as aerospace sealants and linings for fuel tanks. A first preferred polymerizable composition thus includes at least one polythioether as described herein; a curing agent or combination of curing agents; and a filler.

The polythioether or combination of polythioethers preferably is present in the polymerizable composition in an

amount from about 30 wt % to about 90 wt %, more preferably about 40 to about 80 wt %, very preferably about 45 to about 75 wt %, with the wt % being calculated based on the weight of all non-volatile components of the composition. Preferably, the  $T_g$  of the polythioether(s) used in the polymerizable composition is not higher than -55° C., more preferably not higher than -60° C.

Curing agents useful in polymerizable compositions of the invention include epoxy resins, for example, hydantoin diepoxide, diglycidyl ether of bisphenol-A epoxides, diglycidyl ether of bisphenol-F epoxides, Novolak type epoxides, and any of the epoxidized unsaturated and phenolic resins. Other useful curing agents include unsaturated compounds such as acrylic and methacrylic esters of commercially available polyols, unsaturated synthetic or naturally occurring resin compounds, TAC, and olefinic terminated derivatives of the compounds of the present invention. In addition, useful cures can be obtained through oxidative coupling of the thiol groups using organic and inorganic peroxides (e.g.,  $MnO_2$ ) known to those skilled in the art. Selection of the particular curing agent may affect the  $T_g$  of the cured composition. For example, curing agents that have a  $T_g$  significantly lower than the  $T_g$  of the polythioether may lower the  $T_g$  of the cured composition.

Depending on the nature of the polythioether(s) used in the composition, the composition will contain about 90% to about 150% of the stoichiometric amount, preferably about 95 to about 125%, of the selected curing agent(s).

Fillers useful in the polymerizable compositions of the invention include those commonly used in the art, such as carbon black and calcium carbonate ( $CaCO_3$ ). Preferably, the compositions include about 5 to about 60 wt % of the selected filler or combination of fillers, very preferably about 10 to 50 wt %.

The polythioethers, curing agents and fillers employed in polymerizable compositions of the invention, as well as optional additives as described below, should be selected so as to be compatible with each other. Selection of compatible ingredients for the inventive compositions can readily be performed by those skilled in the art without recourse to undue experimentation.

The foregoing polymerizable compositions preferably are curable at a minimum temperature of about 0° C. (i.e., at a temperature of about 0° C. or higher), more preferably about -10° C., very preferably about -20° C., and have a  $T_g$  when cured not higher than about -55° C., more preferably not higher than -60° C., very preferably not higher than -65° C. When cured, the polymerizable compositions preferably have a % volume swell not greater than 25%, more preferably not greater than 20%, after immersion for one week at 60° C. (140° F.) and ambient pressure in jet reference fluid (JRF) type 1.

In addition to the foregoing ingredients, polymerizable compositions of the invention can optionally include one or more of the following: pigments; thixotropes; accelerators; retardants; adhesion promoters; and masking agents.

Useful pigments include those conventional in the art, such as carbon black and metal oxides. Pigments preferably are present in an amount from about 0.1 to about 10 wt %.

Thixotropes, for example silica, are preferably used in an amount from about 0.1 to about 5 wt %.

Accelerators known to the art, such as amines, preferably are present in an amount from about 0.1 to about 5 wt %. Two such useful accelerators are 1,4-diaza-bicyclo[2.2.2]octane (DABCO®), commercially available from Air Products, Chemical Additives Division, Allentown, Pa.) and DMP-30® (an accelerant composition including 2,4,6-tri

(dimethylaminomethyl)phenol, commercially available from Rohm and Haas, Philadelphia, Pa.).

Retardants, such as stearic acid, likewise preferably are used in an amount from about 0.1 to about 5 wt %. Adhesion promoters, which can be, for example, conventional phenolics or silanes, if employed are preferably present in amount from about 0.1 to about 5 wt %. Masking agents, such as pine fragrance or other scents, which are useful in covering any low level odor of the composition, are preferably present in an amount from about 0.1 to about 1 wt %.

An additional advantage of sealant compositions according to the invention is their improved curing behavior. The extent of cure of a sealant composition as a function of time is often difficult to measure directly, but can be estimated by determining the extrusion rate of the composition as a function of time. The extrusion rate is the rate at which a mixed sealant composition, i.e., a sealant composition together with an accelerator system, is extruded from an applicator device. Since the sealant composition is mixed with the accelerator system, curing begins, and the extrusion rate changes with time. The extrusion rate thus is inversely related to the extent of cure. That is, when the extent of cure is low, the viscosity of the mixed sealant composition is low and thus the extrusion rate is high. When the reaction approaches completion, the viscosity becomes very high, and the extrusion rate thus becomes low.

With reference to FIG. 1, the viscosity of some known types sealant compositions remains low for an extended time, because the compositions are slow to cure. Such compositions have extrusion curves qualitatively similar to curve A. Other known types of sealant composition cure very quickly, and thus their viscosity rapidly increases. Consequently, the extrusion rate rapidly decreases, as shown in curve B. Desirably, a mixed sealant composition should have a low viscosity, and thus a high extrusion rate, for a length of time sufficient to allow even application of the sealant composition to the area requiring sealing, but then should cure rapidly after application, i.e., their extrusion rate should quickly decrease. Sealant compositions according to the present invention are characterized by this desirable extrusion curve, as illustrated qualitatively in curve C.

Sealant compositions according to the present invention can have, depending on the particular formulation, initial extrusion rates as high as 500 g/min or higher, together with low extrusion rates on the order of about 5 to 10 g/min or less after curing times on the order of one hour.

As shown in FIG. 2, the initial extrusion rate of a polymer of the present invention (Example 1, below, cured with an epoxy curing agent as described below) is about 550 g/min, then falls rapidly to about 20 g/min after 70 minutes. In comparison, a known polysulfide (cured with  $\text{MnO}_2$ ) has an initial extrusion rate of about 90 g/min, which slowly falls to about 20 g/min after 70 minutes.

A second preferred polymerizable composition combines one or more plasticizers with the polythioether(s), curing agent(s) and filler(s) described above. Use of a plasticizer allows the polymerizable composition to include polythioethers which have higher  $T_g$  than would ordinarily be useful in an aerospace sealant. That is, use of a plasticizer effectively reduces the  $T_g$  of the composition, and thus increases the low-temperature flexibility of the cured polymerizable composition beyond that which would be expected on the basis of the  $T_g$  of the polythioethers alone.

Plasticizers that are useful in polymerizable compositions of the invention include phthalate esters, chlorinated paraffins, hydrogenated terphenyls, etc. The plasticizer or combination of plasticizers preferably constitute 1 to about 40 wt %, more preferably 1 to about 10 wt % of the composition.

Depending on the nature and amount of the plasticizers used in the composition, polythioethers of the invention which have  $T_g$  values up to about  $-50^\circ\text{C}$ ., preferably up to about  $-55^\circ\text{C}$ ., can be used.

The foregoing polymerizable compositions also preferably are curable at a minimum temperature of about  $0^\circ\text{C}$ ., more preferably about  $-10^\circ\text{C}$ ., very preferably about  $-20^\circ\text{C}$ .

The present invention is illustrated in more detail by means of the following non-limiting examples.

In examples 1-8, liquid polythioethers were prepared by stirring together one or more dithiols with one or more divinyl ethers and a trifunctionalizing agent. The reaction mixture was then heated and a free radical catalyst was added. All reactions proceeded substantially to completion (approximately 100% yield).

#### EXAMPLE 1

In a 2 L flask, 524.8 g (3.32 mol) of diethylene glycol divinyl ether (DEG-DVE) and 706.7 g (3.87 mol) of dimercaptodioxaoctane (DMDO) were mixed with 19.7 g (0.08 mol) of triallylcyanurate (TAC) and heated to  $77^\circ\text{C}$ . To the heated reaction mixture was added 4.6 g (0.024 mol) of an azobisisnitrile free radical catalyst (VAZO® 67 [2,2'-azobis(2-methylbutyronitrile), commercially available from DuPont]). The reaction proceeded substantially to completion after 2 hours to afford 1250 g (0.39 mol, yield 100%) of a liquid polythioether resin having a  $T_g$  of  $-68^\circ\text{C}$ . and a viscosity of 65 poise. The resin was faintly yellow and had low odor.

#### EXAMPLE 2

In a 1 L flask, 404.4 g (1.60 mol) of PLURIOL® E-200 divinyl ether and 355.88 g (1.94 mol) of DMDO were mixed with 12.1 g (0.049 mol) of TAC and reacted as in Example 1. The reaction proceeded substantially to completion after 5 hours to afford 772 g (0.024 mol, yield 100%) of a resin having a  $T_g$  of  $-66^\circ\text{C}$ . and a viscosity of 48 poise. The resin was yellow and had low odor.

#### EXAMPLE 3

In a 100 mL flask, 33.2 g (0.21 mol) of DEG-DVE and 26.48 g (0.244 mol) of 1,2-propanedithiol were mixed with 0.75 g (0.003 mol) of TAC and heated to  $71^\circ\text{C}$ . To the heated reaction mixture was added 0.15 g (0.8 mmol) of VAZO® 67. The reaction proceeded substantially to completion after 7 hours to afford 60 g (0.03 mol, yield 100%) of a resin having a  $T_g$  of  $-61^\circ\text{C}$ . and a viscosity of 22 poise. The resin had a noticeable PDT odor.

#### EXAMPLE 4

In a 100 mL flask, 33.3 g (0.136 mol) of tripropylene glycol divinyl ether (DPE-3) and 27.0 g (0.170 mol) of dimercaptodimethylsulfide (DMDS) were mixed with 0.69 g (0.003 mol) of TAC and heated to  $77^\circ\text{C}$ . To the heated reaction mixture was added 0.15 g (0.8 mmol) of VAZO® 67. The reaction proceeded substantially to completion after 6 hours to afford 61 g (0.028 mol, yield 100%) of a resin having a  $T_g$  of  $-63^\circ\text{C}$ . and a viscosity of 26 poise.

#### EXAMPLE 5

In a 250 mL flask, 113.01 g (0.447 mol) of PLURIOL® E-200 divinyl ether and 91.43 g (0.498 mol) of DMDO were mixed with 1.83 g (0.013 mol) of 1,2,3-propanethiol (PTI) and allowed to react exothermically for 72 hours. The

mixture was then heated to 80° C. To the heated reaction mixture was added 0.2 g (1 mmol) of VAZO® 67. The reaction mixture was maintained at 80° C., and the reaction proceeded substantially to completion after 3 hours to afford 200 g (0.06 mol, yield 100%) of a resin having a  $T_g$  of -66° C. and a viscosity of 55 poise.

## EXAMPLE 6

In a small jar, 14.0 g (0.055 mol) of PLURIOL® E-200 divinyl ether, 6.16 g (0.336 mol) of DMDO and 5.38 g (0.336 mol) of DMDS were mixed with 0.42 g (0.017 mol) of TAC (briefly heated to melt the TAC) and heated to 82° C. To the heated reaction mixture was added 0.2 g (0.001 mol) of VAZO® 67. The reaction proceeded substantially to completion after 18 hours to afford 26 g (8.4 mmol, yield 100%) of a resin having a  $T_g$  of -63° C. and a viscosity of 80 poise.

## EXAMPLE 7

In a small jar, 13.55 g (0.054 mol) of PLURIOL® E-200 divinyl ether, 10.44 g (0.057 mol) of DMDO and 1.44 g (8.1 mmol) of ethylcyclohexanedithiol (ECHDT) were mixed with 0.40 g (1.6 mmol) of TAC (heated briefly to melt the TAC) and heated to 82° C. To the heated reaction mixture was added 0.2 g (0.001 mol) of VAZO® 67. The reaction proceeded substantially to completion after 5 hours to afford 26 g (8.1 mmol, yield 100%) of a resin having a  $T_g$  of -66° C. and a viscosity of 58 poise.

## EXAMPLE 8

In a small glass jar, 9.11 g (0.036 mol) of PLURIOL® E-200 divinyl ether, 5.71 g (0.031 mol) of DMDO, 1.52 g (7.8 mmol) of ECHDT, 5.08 g (0.031 mol) of DMDS and 4.11 g (0.024 mol) of hexanediol divinyl ether (HD-DVE) were mixed with 0.39 g (1.6 mmol) of TAC (heated briefly to dissolve the TAC) and heated to 82° C. To the heated reaction mixture was added 0.6 g (3.1 mmol) of VAZO® 67. The reaction proceeded substantially to completion after about 45 hours to afford 2.6 g (7.8 mmol, yield 100%) of a resin having a  $T_g$  of -66° C. and a viscosity of 304 poise. The resin had a cloudy appearance.

Each of the foregoing resins was evaluated for odor. The following scale was employed: 3: strong, offensive odor; 2: moderate odor; 1: slight odor; 0: substantially odorless.

The polymer described in Example 3 of U.S. Pat. No. 4,366,307 was used as a control. This polymer (the "control polymer") had an odor of 3.

Results were as follows:

Polymer	Odor	Polymer	Odor
1	1	5	1
2	1	6	1
3	3	7	1
4	1	8	2

All of the liquid polythioethers thus had little or moderate odor except polymer 3, which had a strong odor.

The resins prepared in Examples 1-8 were then cured. Curing was carried out using the uncompounded resins with a curing agent and DABCO accelerator. The curing agent had the following composition:

epoxy novolak (equivalent weight 175.5)	22 wt %
hydantoin epoxy (equivalent weight 132)	34 wt %
calcium carbonate	34 wt %
carbon black	5 wt %
silane adhesive promoter	5 wt %

The cured resins were evaluated for odor according to the procedure set forth above. The  $T_g$  and the percent weight gain after immersion in JRF type 1 for one week at room temperature and pressure were also measured for each of the cured resins. The volume swell and weight gain percentages were determined for each cured material as follows:

$w_1$  = initial weight in air

$w_2$  = initial weight in  $H_2O$

$w_3$  = final weight in air

$w_4$  = final weight in  $H_2O$

% volume swell =  $100 \times [(w_2 + w_3) - (w_1 + w_4)] / (w_1 - w_2)$

% weight gain =  $100 \times (w_3 - w_1) / w_1$

The results are given in Table 1:

TABLE 1

	Cured Resin							
	1	2	3	4	5	6	7	8
Odor	0	0	0	0	0	0	0	0
$T_g$ (° C.)	-59	-61	-61	-63	-62	-56	-59	-58
% fuel swell	19	22	—	—	23	19	24	27
% wt gain	14	15	15	23	15	15	19	20

In comparison, the control polymer had an odor of 1-2 when cured.

## EXAMPLE 9

Polythioethers having a number average molecular weight of 2100 and an average functionality  $F$  of 2.1 were prepared by combining a divinyl ether with a dithiol as shown in Table 2 and reacting the materials as described herein. The uncompounded polythioethers were then cured using 15 g of the curing agent described above and 0.30 g of DABCO. For each polythioether so prepared, the following quantities were measured: viscosity (uncured material, poise  $p$ ); Shore A hardness (cured material, Rex durometer value); % weight gain (cured material) after one week at 140° F. (60° C.) and atmospheric pressure in JRF type 1; and  $T_g$  (uncured material, ° C.). Results were as follows:

TABLE 2

divinyl ether	dithiol			
	ECHDT	DMDS	DMDO	HDV <sup>a</sup>
DEG-DVE	145 p 44 Rex 27% -53	(solid) 94 Rex 3% -63	27 p 25 Rex 14% -69	24 p 25 Rex 29% -77
PLURIOL® *	77 p 43 Rex 27% -57	41 p 47 Rex 11% -61	59 p 27 Rex 18% -67	25 p 23 Rex 30% -76
BD-DVE <sup>b</sup>	185 p 42 Rex 44% -59	(solid) — — —	(solid) 20 Rex 21% -79	(solid) 22 Rex 44% -85
HD-DVE	155 p 50 Rex	(solid) —	(solid) 14 Rex	(soft solid) 29 Rex

TABLE 2-continued

divinyl ether	dithiol			
	ECHDT	DMDS	DMDO	HDT <sup>d</sup>
	57%	—	27%	68%
	-60	-63	-78	-86
Poly-THF <sup>c</sup>	91 p	(solid)	27 p	—
	30 Rex	75 Rex	17 Rex	—
	64%	29%	37%	—
	-69	-79	-79	—

<sup>a</sup>PLURIOL® E-200 divinyl ether<sup>b</sup>Butanediol divinyl ether<sup>c</sup>Polytetrahydrofuran divinyl ether<sup>d</sup>Hexanedithiol

From the foregoing table it is apparent that the following combinations of divinyl ether and dithiol afford liquid polythioethers having unexpectedly superior fuel resistance and low temperature flexibility when cured: PLURIOL® E-200/DMDO; and DEG-DVE/DMDO. Other potentially useful combinations include DEG-DVE/ECHDT; DEG-DVE/HDT; PLURIOL® E-200/ECHDT; PLURIOL® E-200/HDT; and poly-THF/DMDO. PLURIOL® E-200/DMDS also has excellent fuel resistance and low temperature flexibility when cured, but the uncompounded material does not remain in a liquid state for an extended period of time.

## EXAMPLE 10

Addition of DMDS to PLURIOL®/DMDO Polymers

Four liquid polythiols were prepared as described herein. The polymers had the following compositions (listed values are molar equivalents):

	1	2	3	4
PLURIOL®	6.6	6.6	6.6	6.6
E-200	8	6	4.5	4
DMDO	0	2	3.5	4
DMDS				

Each uncompounded polymer was cured as in Example 9 (15 g of the curing agent composition and 0.30 g of DABCO), with the addition of 0.2 molar equivalents of TAC to afford polymers having a number average molecular weight of about 3000 and a functionality F of 2.2. For each polymer, the following properties were measured:  $T_g$  (resin, ° C.);  $T_g$  (cured, ° C.); viscosity (p); % swell in JRF type 1; % weight gain in JRF type 1; and % weight gain in water. Results are given in Table 3.

TABLE 3

	1	2	3	4
$T_g$ (resin)	-67	-66	-64	-63
(cured)	-59	-58	-56	-56
Viscosity	59	53	62	80
JRF				
% Swell	24	21	21	20
% Wt Gain	18	15	16	16
H <sub>2</sub> O				
% Wt Gain	11.8	11.5	7.4	7.5

All of the foregoing polymers displayed excellent fuel resistance. Polymers 1 and 2 in particular also displayed excellent low temperature flexibility.

## EXAMPLE 11

Addition of ECHDT to PLURIOL®/DMDO Polymers

Four liquid polythiols were prepared as described herein. The polymers had the following compositions (listed values are molar equivalents):

	1	2	3	4
PLURIOL®	6.6	6.6	6.6	6.6
E-200	8	7	6	5
DMDO	0	1	2	3
ECHDT				

Each uncompounded polymer was cured as in Example 10 to afford polymers having a number average molecular weight of about 3000 and a functionality F of 2.2. For each polymer, the following properties were measured:  $T_g$  (resin, ° C.);  $T_g$  (cured, ° C.); viscosity (p); % swell in JRF type 1; and % weight gain in water. Results are given in Table 4.

TABLE 4

	1	2	3	4
$T_g$ (resin)	-67	-66	-65	-64
(cured)	-59	-59	-58	-56
Viscosity	59	36	44	50
JRF type 1				
% Swell	24	25	28	29
% Wt Gain	18	18	19	19
H <sub>2</sub> O				
% Wt Gain	11.8	10.8	8.3	7.8

All of the foregoing polymers displayed good fuel resistance and low temperature flexibility.

## EXAMPLE 12

In a 250 mL 3-neck flask equipped with a stirrer, thermometer and condenser, 87.7 g (0.554 mol) of DEG-DVE and 112.3 g (0.616 mol) of DMDO are mixed and heated to 77° C. (about 170° F.). To the mixture is added 0.8 g (4.2 mmol) of VAZO® 67 catalyst. The reaction mixture is reacted at 82° C. (about 180° F.) for about 6 hours to afford 200 g (0.06 mol, yield 100%) of a low viscosity liquid polythioether resin having a thiol equivalent of 1625 and a functionality F of 2.0.

## EXAMPLE 13

In a 250 mL 3-neck flask equipped with a stirrer, thermometer and condenser, 26.7 g (0.107 mol) of TAC, 56.4 g (0.357 mol) of DEG-DVE and 117.0 g (0.642 mol) of DMDO are mixed and heated to 77° C. (about 170° F.). To the mixture is added 0.8 g (4.2 mmol) of VAZO 67 catalyst. The reaction mixture is reacted at 82° C. (about 180° F.) for about 6 hours to afford 200 g (0.07 mol, yield 100%) of a high viscosity liquid polythioether resin having an equivalent of 800 and a functionality F of about 3.5.

## EXAMPLE 14

Sealant Composition

A sealant composition including the DMDO/DEG-DVE polythioether polymer of Example 1 was compounded as follows (amounts in parts by weight):

DMDO/DEG-DVE Polythioether	100
Calcium carbonate	60
Magnesium oxide	1
Phenolic resin	1
DMP-30	1
Isopropyl alcohol	3

The compounded polymer was mixed intimately with the epoxy resin curing agent of Examples 9–11 above, in the weight ratio of 10:1 and cured at ambient temperature and humidity. The following physical properties were obtained for the cured composition:

Cure hardness at 25° C.	60 Shore A
Tensile strength at break	550 psi
Elongation at break	600%
Notched tear strength	100 p/l
Low-temperature flexibility (AMS 3267 § 4.5.4.7)	passed

#### EXAMPLE 15

##### Sealant Composition

A sealant composition including the ECHDT/DEG-DVE polythioether polymer of Example 9 was compounded as follows (amounts in parts by weight):

ECHDT/DEG-DVE Polythioether	100
Calcium carbonate	54
Hydrated aluminum oxide	20
Magnesium oxide	1
Phenolic resin	1
Hydrogenated terphenyl plasticizer	6
DMP-30	1
Isopropyl alcohol	3

The compounded polymer was mixed intimately with an epoxy resin curing agent in the weight ratio of 10:1 and cured at ambient temperature and humidity. The following physical properties were obtained for the cured composition:

Cure hardness at 25° C.	72 Shore A
Tensile strength at break	550 psi
Elongation at break	450%
Notched tear strength	85 p/l
Low-temperature flexibility	passed

#### EXAMPLE 16

##### OII-Terminated Capped Polythioether

In a 500 ml flask, 275.9 g (1.09 mol) PLURIOL® E-200 divinyl ether, 174.7 g (0.95 mol) DMDO, 28.7 g (0.30 mol) 3-mercaptopropanol and 1.83 g (7.3 mmol) TAC were mixed. The mixture was heated to 70° C., and 2.3 g (12 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85–90° C. for 4 hours to afford 480 g (0.15 mol, yield 100%) of a polymer having an equivalent weight of 1670 (number average molecular weight=3200, functionality F=2.05).

#### EXAMPLE 17

##### OII-Terminated Capped Polythioether

In a 250 ml flask, 104.72 g (0.57 mol) DMDO, 80.73 g (0.51 mol) DEG-DVE and 14.96 g (0.13 mol) butanediol

monovinyl ether were mixed and heated to 75° C. To the heated mixture 0.60 g (3 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 75–85° C. for 6 hours to afford 200 g (0.064 mol, yield 100%) of a clear, nearly colorless polymer with very low odor and a viscosity of 79 poise at 20° C. The equivalent weight was 1570 (number average molecular weight=3200, functionality F=2.00).

#### EXAMPLE 18

##### Vinyl-Terminated Polythioether

In a 250 ml flask, 97.63 g (0.53 mol) DMDO, 97.66 g (0.62 mol) DEG-DVE and 5.31 g (0.21 mol) TAC were mixed and heated to 70° C. To the heated mixture 0.80 g (4 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85–90° C. for 4 hours to afford 200 g (0.11 mol, yield 100%) of a low-odor polymer having a  $T_g$  of -68° C. and a viscosity of 25 poise at 20° C. The equivalent weight was 1570 (number average molecular weight=1900, functionality F=2.2).

#### EXAMPLE 19

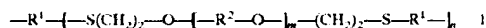
##### Ethyl Vinyl Ether-Terminated Polythioether

In a 100 ml flask, 43.05 g (0.24 mol) DMDO, 34.22 g (0.22 mol) DEG-DVE and 2.84 g (0.04 mol) ethyl vinyl ether were mixed and heated to 80° C. To the heated mixture 0.28 g (1.5 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85° C. for 6 hours to afford 80 g (0.02 mol, yield 100%) of a polymer having a  $T_g$  of -67° C. and a viscosity of 64 poise at 20° C. (number average molecular weight=4100, functionality F=2.0).

What is claimed is:

1. A polymerizable composition comprising

(i) about 30 to about 90 wt % of at least one polythioether, said at least one polythioether comprising a structure having the formula 1



wherein

$R^1$  denotes a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-10}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $-[(CH_2)_p-X]_q-(CH_2)_r-$ , or  $-[(CH_2)_p-X]_q-(CH_2)_r-$  in which at least one  $-CH_2-$  unit is substituted with a methyl group,

$R^2$  denotes methylene, a divalent  $C_{2-6}$  n-alkyl,  $C_{2-6}$  branched alkyl,  $C_{6-10}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group, or  $-[(CH_2)_p-X]_q-(CH_2)_r-$ ,

X is one selected from the group consisting of O, S and  $-NR^6-$ .

$R^6$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10,

said at least one polythioether being a liquid at room temperature and pressure,

said at least one polythioether having a glass transition temperature not higher than -55° C,

(ii) a curing agent in an amount from about 90 to about 150% of stoichiometric based on the amount of at least one polythioether, and

(iii) about 5 to about 60 wt % of a filler, with all wt % being based on the total weight of non-volatile components of the composition,

wherein said composition is curable at a minimum temperature of 0 C.

2. The polymerizable composition of claim 1 which has a glass transition temperature  $T_g$  not higher than  $-60^\circ\text{C}$ .

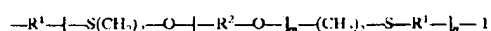
3. The polymerizable composition of claim 1 which, when cured, has a percent volume swell not greater than 25% after immersion in JRF type 1 for one week at  $60^\circ\text{C}$ . and ambient pressure.

4. The polymerizable composition of claim 1 further comprising an additive selected from the group consisting of a pigment in an amount from about 0.1 to about 10 wt %, a thixotrope in an amount from about 0.1 to about 5 wt %, an accelerator in an amount from about 0.1 to about 5 wt %, a retardant in an amount from about 0.1 to about 5 wt %, an adhesion promoter in an amount from about 0.1 to about 5 wt %, and a masking agent in an amount from about 0.1 to about 1 wt %.

5. The polymerizable composition of claim 1 which comprises a mixture of at least two different polythioethers (i).

6. A polymerizable composition comprising

(i) about 30 to about 90 wt % of at least one polythioether, said at least one polythioether comprising a structure having the formula 1



wherein

$R^1$  denotes a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $-[(CH_2)_p-X]_q-(CH_2)_r-$ , or  $-[(CH_2)_p-X]_q-(CH_2)_r-$  in which at least one  $-CH_2-$  unit is substituted with a methyl group,

$R^2$  denotes methylene, a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group, or  $-[(CH_2)_p-X]_q-(CH_2)_r-$ ,

X is one selected from the group consisting of O, S and  $-NR^6-$ ,

$R^6$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10,

said at least one polythioether being a liquid at room temperature and pressure, said at least one polythioether having a glass transition temperature not greater than  $-50^\circ\text{C}$ ,

(ii) a curing agent in an amount from about 90 to about 150% of stoichiometric based on the amount of at least one polythioether,

(iii) a plasticizer in an amount from about 1 to about 40 wt %, and

(iv) about 5 to about 60 wt % of a filler, with all wt % being based on the total weight of non-volatile components of the composition,

wherein said composition is curable at a minimum temperature of 0 C.

7. The polymerizable composition of claim 6 which has a glass transition temperature  $T_g$  not greater than  $-55^\circ\text{C}$ .

8. The polymerizable composition of claim 6 which, when cured, has a percent volume swell not greater than 25% after immersion for one week at room temperature and pressure.

9. The polymerizable composition of claim 6 wherein said plasticizer is selected from the group consisting of phthalate esters, chlorinated paraffins and hydrogenated terphenyls.

10. The polymerizable composition of claim 6 further comprising an additive selected from the group consisting of a pigment in an amount from about 0.1 to about 10 wt %, a thixotrope in an amount from about 0.1 to about 5 wt %, an accelerator in an amount from about 0.1 to about 5 wt %, a retardant in an amount from about 0.1 to about 5 wt %, an adhesion promoter in an amount from about 0.1 to about 5 wt %, and a masking agent in an amount from about 0.1 to about 1 wt %.

11. The polymerizable composition of claim 6 which comprises a mixture of at least two different polythioethers (i).

\* \* \* \* \*



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**OCT 19 2007**

**OFFICE OF PETITIONS**

In re Application of  
Jonathan D. ZOOK et al  
Application No. 10/302,177  
Filed: November 21, 2002  
Attorney Docket No. 8303.0042-05

:  
:  
: DECISION ON PETITION  
:  
:

This is a decision on the petition under the unintentional provisions of 37 CFR 1.137(b), filed October 9, 2007, to revive the above-identified application.

The petition is **GRANTED**.

The application became abandoned for failure to reply in a timely manner to the non-final Office action mailed February 19, 2003, which set a shortened statutory period for reply of three (3) months. No extensions of time under the provisions of 37 CFR 1.136(a) were obtained. Accordingly, the application became abandoned on May 20, 2003. A Notice of Abandonment was mailed on September 15, 2003.

A grantable petition under 37 CFR 1.137(b) must be accompanied by:

- (1) the required reply, unless previously filed;
- (2) the petition fee as set forth in 37 CFR 1.17(m);
- (3) a statement that the entire delay in filing the required reply from the due date for the reply until the filing of a grantable petition pursuant to 37 CFR 1.137(b) was unintentional; and
- (4) any terminal disclaimer (and fee as set forth in 37 CFR 1.20(d)) required by 37 CFR 1.137(d).

The petition satisfies the requirements of 37 CFR 1.137(b) in that petitioner has supplied (1) the reply in the form of a continuing application under 37 CFR 1.53(b); (2) the petition fee of \$1,540; and (3) an adequate showing that the entire delay was


**ZOOK EXHIBIT 1031**  
**Zook v. Zook**  
**Interference No. 105,555 (MPT)**

unintentional. No terminal disclaimer is required as a condition for reviving this application.

This application is being revived solely for purposes of continuity. Therefore, as continuity has been established by this decision reviving this application, this application is again abandoned in favor of continuing Application No. 10/644,389, filed August 19, 2003.

As authorized, the \$1,540 petition fee required by 37 CFR 1.17(m) will be charged to petitioner's deposit account. Although authorized, no fee will be charged to an extension of time. An extension of time under 37 CFR 1.136 must be filed prior to the expiration of the maximum extendable period for reply. See In re Application of S, 8 USPQ2d 1630, 1631 (Comm'r Pats. 1988).

Telephone inquiries concerning this decision should be directed to the undersigned at (571) 272-3218.

  
Frances Hicks  
Petitions Examiner  
Office of Petitions

**UTILITY PATENT APPLICATION TRANSMITTAL**  
**(Large Entity)***(Only for new nonprovisional applications under 37 CFR 1.53(b))*Docket No.  
OC-573A1Total Pages in this Submission  
48**TO THE ASSISTANT COMMISSIONER FOR PATENTS**Box Patent Application  
Washington, D.C. 20231

Transmitted herewith for filing under 35 U.S.C. 111(a) and 37 C.F.R. 1.53(b) is a new utility patent application for an invention entitled:

**SEALANTS AND POTTING FORMULATIONS INCLUDING MERCAPTO-TERMINATED POLYMERS  
PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER**

and invented by:

**Jonathan D. Zook, Suzanna G. DeMoss, David W. Jordan, Chandra B. Rao, Hakam Singh and  
Ahmed Sharaby**If a **CONTINUATION APPLICATION**, check appropriate box and supply the requisite information:☐ Continuation ☐ Divisional ☒ Continuation-in-part (CIP) of prior application No.: 08/928,972

Which is a:

☐ Continuation ☐ Divisional ☒ Continuation-in-part (CIP) of prior application No.: 08/802,130

Which is a:

☐ Continuation ☐ Divisional ☒ Continuation-in-part (CIP) of prior application No.: 09/318,500

Enclosed are:

**Application Elements**

1. ☒ Filing fee as calculated and transmitted as described below
2. ☒ Specification having 40 pages and including the following:
  - a. ☒ Descriptive Title of the Invention
  - b. ☒ Cross References to Related Applications *(if applicable)*
  - c. ☐ Statement Regarding Federally-sponsored Research/Development *(if applicable)*
  - d. ☐ Reference to Microfiche Appendix *(if applicable)*
  - e. ☒ Background of the Invention
  - f. ☒ Brief Summary of the Invention
  - g. ☒ Brief Description of the Drawings *(if drawings filed)*
  - h. ☒ Detailed Description
  - i. ☒ Claim(s) as Classified Below
  - j. ☒ Abstract of the Disclosure

**UTILITY PATENT APPLICATION TRANSMITTAL**  
**(Large Entity)**

*(Only for new nonprovisional applications under 37 CFR 1.53(b))*

Docket No.  
OC-573A1

Total Pages in this Submission  
48

**Application Elements (Continued)**

3. ☒ Drawing(s) *(when necessary as prescribed by 35 USC 113)*
- a. ☐ Formal                      Number of Sheets \_\_\_\_\_
- b. ☒ Informal                      Number of Sheets                      2
4. ☒ Oath or Declaration
- a. ☐ Newly executed *(original or copy)*                      ☒ Unexecuted
- b. ☐ Copy from a prior application (37 CFR 1.63(d)) *(for continuation/divisional application only)*
- c. ☒ With Power of Attorney                      ☐ Without Power of Attorney
- d. ☐ DELETION OF INVENTOR(S)  
Signed statement attached deleting inventor(s) named in the prior application,  
see 37 C.F.R. 1.63(d)(2) and 1.33(b).
5. ☐ Incorporation By Reference *(usable if Box 4b is checked)*  
The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied  
under Box 4b, is considered as being part of the disclosure of the accompanying application and is hereby  
incorporated by reference therein.
6. ☐ Computer Program in Microfiche *(Appendix)*
7. ☐ Nucleotide and/or Amino Acid Sequence Submission *(if applicable, all must be included)*
- a. ☐ Paper Copy
- b. ☐ Computer Readable Copy *(identical to computer copy)*
- c. ☐ Statement Verifying Identical Paper and Computer Readable Copy

**Accompanying Application Parts**

8. ☐ Assignment Papers *(cover sheet & document(s))*
9. ☐ 37 CFR 3.73(B) Statement *(when there is an assignee)*
10. ☐ English Translation Document *(if applicable)*
11. ☐ Information Disclosure Statement/PTO-1449                      ☐ Copies of IDS Citations
12. ☐ Preliminary Amendment
13. ☒ Acknowledgment postcard
14. ☒ Certificate of Mailing
- ☐ First Class                      ☐ Express Mail *(Specify Label No.):* EL 733 075 345 US

**UTILITY PATENT APPLICATION TRANSMITTAL**  
**(Large Entity)**

*(Only for new nonprovisional applications under 37 CFR 1.53(b))*

Docket No.  
OC-573A1

Total Pages in this Submission  
48

**Accompanying Application Parts (Continued)**

15. ☐ Certified Copy of Priority Document(s) *(if foreign priority is claimed)*
16. ☐ Additional Enclosures *(please identify below):*

**Request That Application Not Be Published Pursuant To 35 U.S.C. 122(b)(2)**

17. ☐ Pursuant to 35 U.S.C. 122(b)(2), Applicant hereby requests that this patent application not be published pursuant to 35 U.S.C. 122(b)(1). Applicant hereby certifies that the invention disclosed in this application has not and will not be the subject of an application filed in another country, or under a multilateral international agreement, that requires publication of applications 18 months after filing of the application.

**Warning**

***An applicant who makes a request not to publish, but who subsequently files in a foreign country or under a multilateral international agreement specified in 35 U.S.C. 122(b)(2)(B)(i), must notify the Director of such filing not later than 45 days after the date of the filing of such foreign or international application. A failure of the applicant to provide such notice within the prescribed period shall result in the application being regarded as abandoned, unless it is shown to the satisfaction of the Director that the delay in submitting the notice was unintentional.***

**UTILITY PATENT APPLICATION TRANSMITTAL  
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Docket No.  
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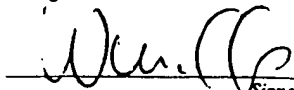
Total Pages in this Submission  
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**Fee Calculation and Transmittal**

**CLAIMS AS FILED**

For	#Filed	#Allowed	#Extra	Rate	Fee
Total Claims	21	- 20 =	1	x \$18.00	\$18.00
Indep. Claims	4	- 3 =	1	x \$80.00	\$80.00
Multiple Dependent Claims (check if applicable) <input type="checkbox"/>					\$0.00
BASIC FEE					\$710.00
OTHER FEE (specify purpose) _____					\$0.00
TOTAL FILING FEE					\$808.00

- ☐ A check in the amount of \_\_\_\_\_ to cover the filing fee is enclosed.
- ☒ The Commissioner is hereby authorized to charge and credit Deposit Account No. 16-2025 as described below. A duplicate copy of this sheet is enclosed.
- ☒ Charge the amount of \$808.00 as filing fee.
  - ☒ Credit any overpayment.
  - ☒ Charge any additional filing fees required under 37 C.F.R. 1.16 and 1.17.
  - ☐ Charge the issue fee set in 37 C.F.R. 1.18 at the mailing of the Notice of Allowance, pursuant to 37 C.F.R. 1.311(b).

  
Signature

**WILLIAM E. KUSS**  
Registration No. 41,919  
Attorney for Applicant

Pittsburgh, PA

Dated: January 8, 2001

CC:

# UTILITY PATENT APPLICATION TRANSMITTAL

## (Large Entity)

(Only for new nonprovisional applications under 37 CFR 1.53(b))

Docket No.  
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and invented by:

**Jonathan D. Zook, Suzanna G. DeMoss, David W. Jordan, Chandra B. Rao, Hakam Singh and  
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**UTILITY PATENT APPLICATION TRANSMITTAL**  
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*(Only for new nonprovisional applications under 37 CFR 1.53(b))*

Docket No.  
OC-573A1

Total Pages in this Submission  
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**Application Elements (Continued)**

3. ☒ Drawing(s) (when necessary as prescribed by 35 USC 113)

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- b. ☒ Informal                      Number of Sheets 2

4. ☒ Oath or Declaration

- a. ☐ Newly executed (original or copy)      ☒ Unexecuted
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- d. ☐ DELETION OF INVENTOR(S)  
Signed statement attached deleting inventor(s) named in the prior application,  
see 37 C.F.R. 1.63(d)(2) and 1.33(b).

5. ☐ Incorporation By Reference (usable if Box 4b is checked)

The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under Box 4b, is considered as being part of the disclosure of the accompanying application and is hereby incorporated by reference therein.

6. ☐ Computer Program in Microfiche (Appendix)

7. ☐ Nucleotide and/or Amino Acid Sequence Submission (if applicable, all must be included)

- a. ☐ Paper Copy
- b. ☐ Computer Readable Copy (identical to computer copy)
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Docket No.  
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Total Pages in this Submission  
48

**Accompanying Application Parts (Continued)**

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**UTILITY PATENT APPLICATION TRANSMITTAL**  
**(Large Entity)**

*(Only for new nonprovisional applications under 37 CFR 1.53(b))*

Docket No.  
**OC-573A1**

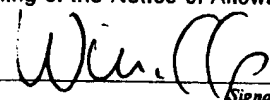
Total Pages in this Submission  
**48**

**Fee Calculation and Transmittal**

**CLAIMS AS FILED**

For	#Filed	#Allowed	#Extra	Rate	Fee
Total Claims	21	- 20 =	1	x \$18.00	\$18.00
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Multiple Dependent Claims (check if applicable) <input type="checkbox"/>					\$0.00
BASIC FEE					\$710.00
OTHER FEE (specify purpose)					\$0.00
TOTAL FILING FEE					\$808.00

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- ☒ Credit any overpayment.
- ☒ Charge any additional filing fees required under 37 C.F.R. 1.16 and 1.17.
- ☐ Charge the issue fee set in 37 C.F.R. 1.18 at the mailing of the Notice of Allowance, pursuant to 37 C.F.R. 1.311(b).

  
Signature

**WILLIAM E. KUSS**  
Registration No. 41,919  
Attorney for Applicant

Pittsburgh, PA

Dated: January 8, 2001

CC:

# United States Patent [19]

Zook et al.

[11] Patent Number: 5,912,319

[45] Date of Patent: Jun. 15, 1999

[54] COMPOSITIONS AND METHOD FOR PRODUCING FUEL RESISTANT LIQUID POLYTHIOETHER POLYMERS WITH GOOD LOW TEMPERATURE FLEXIBILITY

[75] Inventors: Jonathan Doherty Zook, Santa Clarita; Suzanna Gibson, Los Angeles; David Weldon Jordan, Reseda; Chandra B. Rao, Valencia, all of Calif.

[73] Assignee: Courtaulds Aerospace, Inc., Glendale, Calif.

[21] Appl. No.: 08/802,130

[22] Filed: Feb. 19, 1997

[51] Int. Cl.<sup>6</sup> C08G 75/00

[52] U.S. Cl. 528/373; 528/374; 528/378; 525/212; 568/29

[58] Field of Search 528/373, 374, 528/378; 525/212; 568/29

## [56] References Cited

### U.S. PATENT DOCUMENTS

3,506,626 4/1970 Warner .  
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3,725,229 4/1973 Kehr .  
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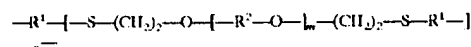
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(List continued on next page.)

Primary Examiner—Duc Truong

## [57] ABSTRACT

A polythioether includes a structure having the formula I



wherein

R<sup>1</sup> denotes a divalent C<sub>2-6</sub> n-alkyl, C<sub>3-6</sub> branched alkyl, C<sub>6-8</sub> cycloalkyl or C<sub>6-10</sub> alkylcycloalkyl group,  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$ , or  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$  in which at least one  $\text{---CH}_2\text{---}$  unit is substituted with a methyl group,

R<sup>2</sup> denotes methylene, a divalent C<sub>2-6</sub> n-alkyl, C<sub>2-6</sub> branched alkyl, C<sub>6-8</sub> cycloalkyl or C<sub>6-10</sub> alkylcycloalkyl group,  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$ , or  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$  in which at least one  $\text{---CH}_2\text{---}$  unit is substituted with a methyl group,

X denotes one selected from the group consisting of O, S and  $\text{---NR}^6$ ,

R<sup>6</sup> denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

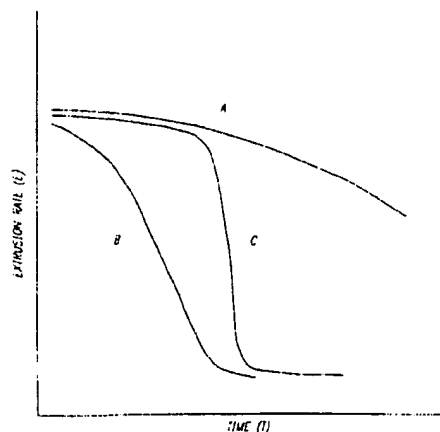
p is an integer from 2 to 6,

q is an integer from 0 to 5, and

r is an integer from 2 to 10.

The polythioether is a liquid at room temperature and pressure.

23 Claims, 2 Drawing Sheets



ZOOK EXHIBIT 1033  
Zook v. Zook  
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FIG. 1

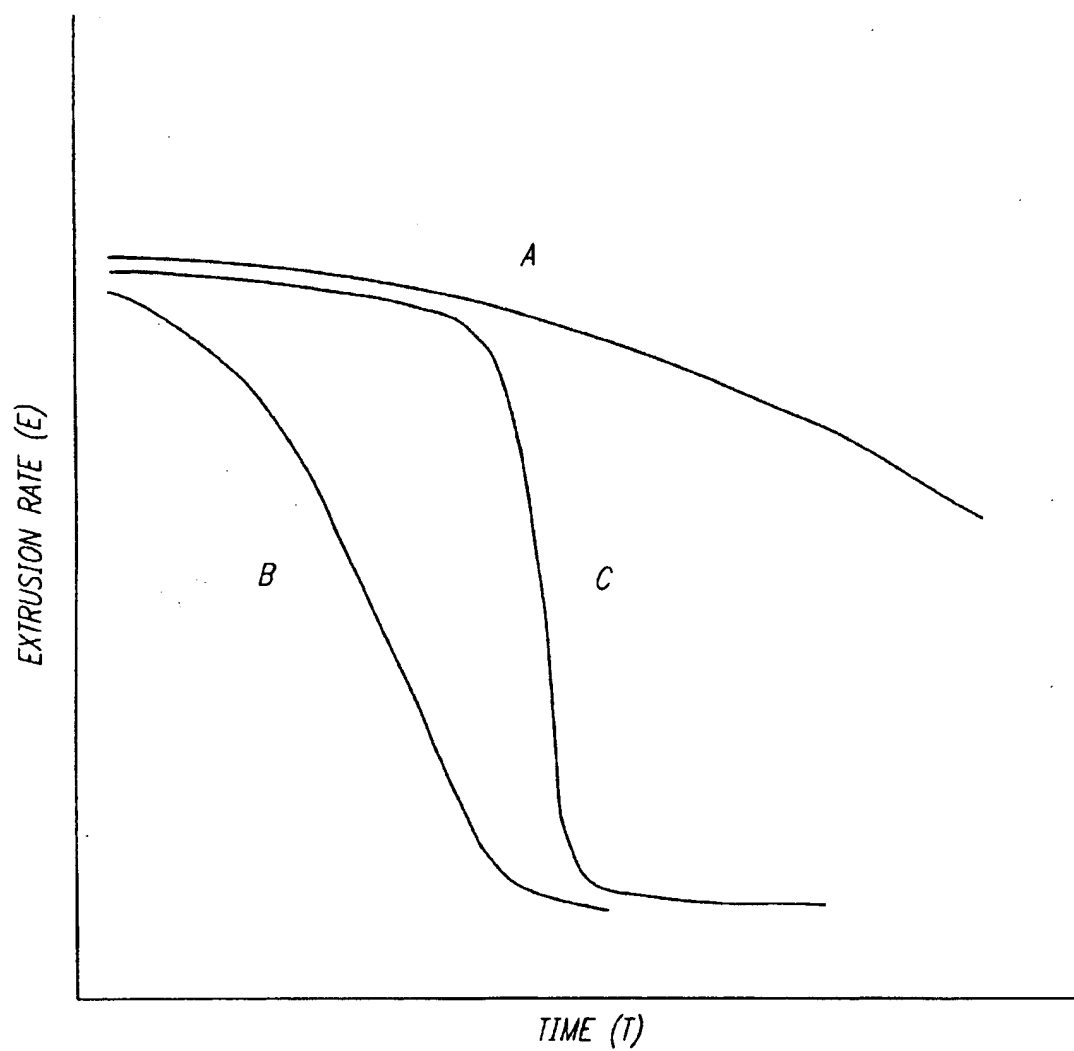
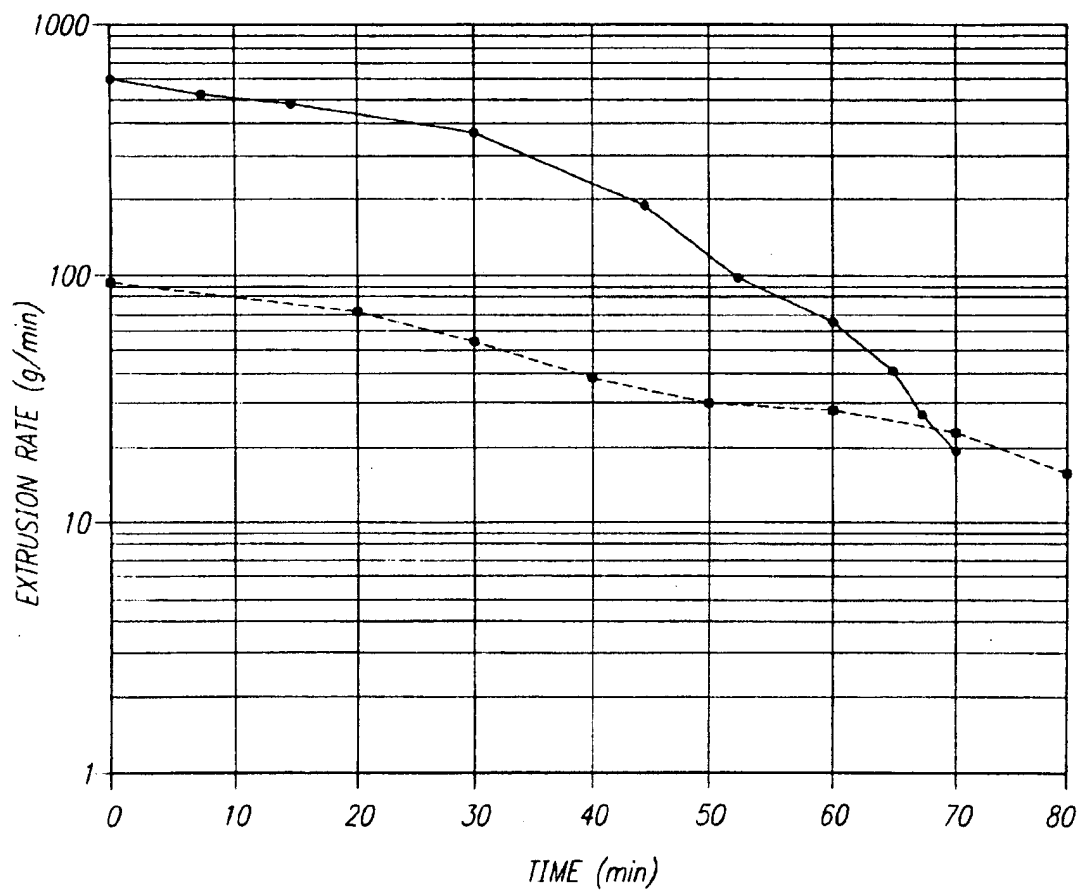


FIG. 2



# COMPOSITIONS AND METHOD FOR PRODUCING FUEL RESISTANT LIQUID POLYTHIOETHER POLYMERS WITH GOOD LOW TEMPERATURE FLEXIBILITY

## FIELD OF THE INVENTION

The present invention relates to liquid polythioether polymers that have good low temperature flexibility and fuel resistance when cured. The invention is also directed to methods for making the polymers by reacting polythiols with oxygenated dienes (divinyl ethers) which substantially eliminate malodorous condensed cyclic by-products.

## BACKGROUND OF THE INVENTION

Thiol-terminated sulfur-containing polymers are known to be well-suited for use in aerospace sealants due to their fuel resistant nature upon cross-linking. Among the commercially available polymeric materials which have sufficient sulfur content to exhibit this desirable property are the polysulfide polyformal polymers described, e.g., in U.S. Pat. No. 2,466,963, and the alkyl side chain containing polythioether polyether polymers described, e.g., in U.S. Pat. No. 4,366,307 to Singh et al. Materials useful in this context also have the desirable properties of low temperature flexibility (low glass transition temperature  $T_g$ ) and liquidity at room temperature.

An additional desirable combination of properties for aerospace sealants which is much more difficult to obtain is the combination of long application time (i.e., the time during which the sealant remains usable) and short curing time (the time required to reach a predetermined strength). Singh et al., U.S. Pat. No. 4,366,307, disclose such materials. Singh et al. teach the acid-catalyzed condensation of hydroxyl-functional thioethers. The hydroxyl groups are in the  $\beta$ -position with respect to a sulfur atom for increased condensation reactivity. The Singh et al. patent also teaches the use of hydroxyl-functional thioethers with pendent methyl groups to afford polymers having good flexibility and liquidity. However, the disclosed condensation reaction has a maximum yield of about 75% of the desired condensation product. Furthermore, the acid-catalyzed reaction of  $\beta$ -hydroxysulfide monomers yields significant quantities (typically not less than about 25%) of an aqueous solution of thermally stable and highly malodorous cyclic byproducts, such as 1-thia-4-oxa-cyclohexane. As a result, the commercial viability of the disclosed polymers is limited.

Another desirable feature in polymers suitable for use in aerospace sealants is high temperature resistance. Inclusion of covalently bonded sulfur atoms in organic polymers has been shown to enhance high temperature performance. However, in the polysulfide polyformal polymers disclosed in U.S. Pat. No. 2,466,963, the multiple  $-S-S-$  linkages in the polymer backbones result in compromised thermal resistance. In the polymers of Singh et al., U.S. Pat. No. 4,366,307, enhanced thermal stability is achieved through replacement of polysulfide linkages with polythioether ( $-S-$ ) linkages. In practice, however, the disclosed materials also have compromised thermal resistance due to traces of the residual acid condensation catalyst.

Morris et al., U.S. Pat. No. 4,609,762, describes reacting dithiols with secondary or tertiary alcohols to afford liquid polythioethers having no oxygen in the polymeric backbone. Cured polymeric materials formed from these polymers have the disadvantage, however, of reduced fuel resistance due to the large number of pendent methyl groups that are present. In addition, residual catalyst from the disclosed process generates undesirable aqueous acidic waste.

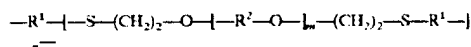
Cameron, U.S. Pat. No. 5,225,472, discloses production of polythioether polymers by the acid-catalyzed condensa-

tion of dithiols with active carbonyl compounds such as  $HCOOH$ . Again, this process generates undesirable aqueous acidic waste.

The addition polymerization of aliphatic dithiols with diene monomers has been described in the literature. See, e.g., Klemm, E. et al., *J. Macromol. Sci.-Chem.*, A28(9), pp. 875-883 (1991); Nuyken, O. et al., *Makromol. Chem., Rapid Commun.* 11, 365-373 (1990). However, neither Klemm et al. nor Nuyken suggest selection of particular starting materials, specifically divinyl ethers and dithiols, such that a polymer is formed that is liquid at room temperature and, upon curing, has excellent low-temperature flexibility (low  $T_g$ ) and high resistance to fuels, i.e., hydrocarbon fluids. Nor do Klemm et al. suggest production of a polymer that in addition is curable at room or lower temperatures. Moreover, the reactions disclosed by Klemm et al. also generate undesirable cyclic byproducts.

## SUMMARY OF THE PREFERRED EMBODIMENTS

In accordance with one aspect of the present invention, there is provided a polythioether having the formula I



wherein

$R^1$  denotes a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $-(CH_2)_p-X-$  or  $-(CH_2)_p-X-$  in which at least one  $-CH_2-$  unit is substituted with a methyl group,

$R^2$  denotes methylene, a divalent  $C_{2-6}$  n-alkyl,  $C_{2-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $-(CH_2)_p-X-$  or  $-(CH_2)_p-X-$  in which at least one  $-CH_2-$  unit is substituted with a methyl group, X denotes one selected from the group consisting of O, S and  $-NR^6-$ ,

$R^6$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10,

the polythioether being a liquid at room temperature and pressure.

Preferably the polythioether has a number average molecular weight between about 500 and about 20,000.

In a first preferred embodiment, the polythioether has the formula II



wherein

A denotes a structure having the formula 1,

y is 0 or 1,

$R^3$  denotes a single bond when  $y=0$  and  $-S-(CH_2)_2-[O-R^2]_n-O-$  when  $y=1$ ,

$R^4$  denotes  $-SH$  or  $-S-(CH_2)_2-O-R^5$  when  $y=0$  and  $-CH_2=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,

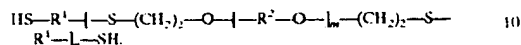
$R^5$  denotes  $C_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $-OH$  or  $-NHR^7$  group, and

$R^7$  denotes H or a  $C_{1-6}$  n-alkyl group.

Polythioethers in which  $R^4$  is  $-SH$  are "uncapped," that is, include unreacted terminal thiol groups. Polythioethers

according to the invention also include "capped" polythioethers, that is, polythioethers including terminal groups other than unreacted thiol groups. These terminal groups can be groups such as —OH or —NH<sub>2</sub>, or groups such as alkyl or terminal ethylenically unsaturated groups.

In a more particular preferred embodiment,  $y=0$  in formula II and  $R^4$  denotes —SH. That is, the polythioether is an uncapped polythioether having the structure

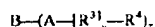


In another more particular preferred embodiment, the inventive polythioether is a capped polythioether in which  $y=0$  in formula II and  $R^4$  denotes  $-S-(CH_2)_2-O-R^5$ . Particularly preferably,  $R^5$  is an unsubstituted or substituted *n*-alkyl group such as ethyl, 4-hydroxybutyl or 3-aminopropyl.

In still another particular preferred embodiment,  $y=1$  in formula II and  $R^4$  denotes  $-CH=CH_2$ . That is, the polythioether is an uncapped polythioether having terminal vinyl groups.

In yet another more particular preferred embodiment, the inventive polythioether is a capped polythioether in which  $y=1$  in formula II and  $R^4$  denotes  $-(CH_2)_2-S-R^5$ .

In a second preferred embodiment, the polythioether has the formula III



wherein

A denotes a structure having the formula 1,

$y$  is 0 or 1,

$R^3$  denotes a single bond when  $y=0$  and  $-S-(CH_2)_2-[-O-R^2]_m-O-$  when  $y=1$ ,

$R^4$  denotes —SH or  $-S-(CH_2)_2-O-R^5$  when  $y=0$  and  $-CH_2=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,

$R^5$  denotes *C*<sub>1-6</sub> *n*-alkyl which is unsubstituted or substituted with at least one —OH or —NHR<sup>7</sup> group,

$R^7$  denotes H or a *C*<sub>1-6</sub> *n*-alkyl group.

$z$  is an integer from 3 to 6, and

B denotes a  $z$ -valent residue of a polyfunctionalizing agent.

That is, the polyfunctionalized embodiments include three or more structures of the formula I bound to the residue of an appropriate polyfunctionalizing agent.

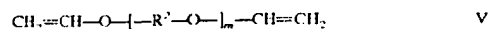
In a more specific embodiment,  $z$  is 3, and the polyfunctionalizing agent thus is a trifunctionalizing agent. In another more specific embodiment, the average functionality of the polythioether ranges between about 2.05 and about 3.00.

In accordance with another aspect of the present invention, there are provided methods of producing the foregoing polythioethers.

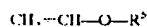
According to a first preferred embodiment, a polythioether of the invention is produced by reacting  $(n+1)$  equivalents (e.g., moles) of a compound having the formula IV



or a mixture of at least two different compounds having the formula IV, with  $(n)$  equivalents of a compound having the formula V



or a mixture of at least two different compounds having the formula V, and optionally about 0.05 to about 2 equivalents of a compound having the formula VI



or a mixture of two different compounds having the formula VI, in the presence of a catalyst. The catalyst is selected from the group consisting of free-radical catalysts, ionic catalysts and ultraviolet light. Preferably the catalyst is a free-radical catalyst such as an azo compound.

According to a second preferred embodiment, a polythioether of the invention is produced by reacting  $(n)$  equivalents of a compound having the formula IV, or a mixture of at least two different compounds having the formula IV, with  $(n+1)$  equivalents of a compound having the formula V, or a mixture of at least two different compounds having the formula V, optionally together with 0.05 to about 2 equivalents of a compound having the formula VII



or a mixture of two different compounds having the formula VII, in the presence of a catalyst as described above.

Analogous methods for producing polyfunctional polythioethers using the foregoing reactants together with appropriate polyfunctionalizing agents are also provided.

Polythioethers produced by the foregoing methods are also provided.

In accordance with yet another aspect of the present invention, there is provided a polymerizable composition comprising (i) about 30 to about 90 wt % of at least one polythioether as defined herein, said at least one polythioether having a glass transition temperature not greater than  $-55^\circ\text{C}$ ., (ii) a curing agent in an amount from about 90 to about 150% of stoichiometric based on the amount of said at least one polythioether, and (iii) about 5 to about 60 wt % of a filler, with all wt % being based on the total weight of non-volatile components of the composition. The inventive composition is curable at a temperature of  $0^\circ\text{C}$ . or higher, preferably at a temperature of  $-20^\circ\text{C}$ . or higher.

In accordance with an additional aspect of the present invention, there is provided a polymerizable composition comprising (i) about 30 to about 90 wt % of at least one polythioether as defined herein, said at least one polythioether having a glass transition temperature not greater than  $-50^\circ\text{C}$ ., (ii) a curing agent in an amount from about 90 to about 150% of stoichiometric based on the amount of said at least one polythioether, (iii) a plasticizer in an amount from about 1 to about 40 wt %, and (iv) a filler in an amount from about 5 to about 60 wt %, with all wt % being based on the total weight of non-volatile components of the composition. The composition is curable at a temperature of  $0^\circ\text{C}$ . or higher, preferably at a temperature of  $-20^\circ\text{C}$ . or higher.

Cured polymeric materials prepared by polymerization of the foregoing compositions are also provided.

Other objects, features and advantages of the present invention will become apparent to those skilled in the art from the following detailed description. It is to be understood, however, that the detailed description and specific examples, while indicating preferred embodiments of the present invention, are given by way of illustration and not limitation. Many changes and modifications within the scope of the present invention may be made without departing from the spirit thereof, and the invention includes all such modifications.

#### BRIEF DESCRIPTION OF THE DRAWINGS

The invention may be more readily understood by referring to the accompanying drawings in which

FIG. 1 depicts linear graphs of extrusion rate (*E*) versus time (*T*) for sealant compositions of the invention in com-

parison to extrusion rate curves for known types of sealant composition, and

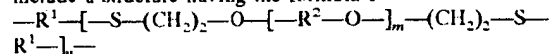
FIG. 2 is a semi-log graph of the extrusion rate curve of a polythioether of the invention (♦) and a prior art polysulfide (■).

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

It has surprisingly been discovered that the combination of certain polythiols with oxygenated dienes according to the present invention results in polythioether polymers that are liquids at room temperature and pressure and that have desirable physical and rheological properties, and that furthermore are substantially free of malodorous cyclic by-products. The inventive materials are also substantially free of deleterious catalyst residues, and hence have superior thermal resistance properties.

According to the present invention, polythioethers are provided that are liquid at room temperature and pressure and have excellent low temperature flexibility (low  $T_g$ ) and fuel resistance. As used herein, the term "room temperature and pressure" denotes approximately 77° F. (25° C.), and 1 atmosphere.

In their most general aspect, the inventive polythioethers include a structure having the formula I



wherein

$\text{R}^1$  denotes a divalent  $\text{C}_{2-6}$  n-alkyl,  $\text{C}_{3-6}$  branched alkyl,  $\text{C}_{6-8}$  cycloalkyl or  $\text{C}_{6-10}$  alkylcycloalkyl group,  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}(\text{—CH}_2\text{—})_r\text{—}$ , or  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}(\text{—CH}_2\text{—})_r\text{—}$  in which at least one  $\text{—CH}_2\text{—}$  unit is substituted with a methyl group,

$\text{R}^2$  denotes methylene, a divalent  $\text{C}_{2-6}$  n-alkyl,  $\text{C}_{2-6}$  branched alkyl,  $\text{C}_{6-8}$  cycloalkyl or  $\text{C}_{6-10}$  alkylcycloalkyl group,  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}(\text{—CH}_2\text{—})_r\text{—}$ , or  $\text{—}[(\text{—CH}_2\text{—})_p\text{—X—}]_q\text{—}(\text{—CH}_2\text{—})_r\text{—}$  in which at least one  $\text{—CH}_2\text{—}$  unit is substituted with a methyl group,

X denotes one selected from the group consisting of O, S and  $\text{—NR}^6\text{—}$ ,

$\text{R}^6$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10.

Preferably, a polythioether polymer according to the invention has a glass transition temperature  $T_g$  that is not higher than  $-50^\circ\text{C}$ . More preferably, the  $T_g$  of the inventive polymer is not higher than  $-55^\circ\text{C}$ . Very preferably, the  $T_g$  of the inventive polymer is not higher than  $-60^\circ\text{C}$ . Low  $T_g$  is indicative of good low temperature flexibility, which can be determined by known methods, for example, by the methods described in AMS (Aerospace Material Specification) 3267 §4.5.4.7, MIL-S (Military Specification) -8802E §3.3.12 and MIL-S-29574, and by methods similar to those described in ASTM (American Society for Testing and Materials) D522-88.

The polythioethers of the invention exhibit very desirable fuel resistance characteristics when cured. One measure of the fuel resistance of the inventive polymers is their percent volume swell after prolonged exposure to a hydrocarbon fuel, which can be quantitatively determined using methods

similar to those described in ASTM D792 or AMS 3269. Thus, in a preferred embodiment, the inventive polymers have, when cured, a percent volume swell not greater than 25% after immersion for one week at 140° F. (60° C.) and ambient pressure in jet reference fluid (JRF) type 1. Very preferably, the percent volume swell of the cured polymers is not greater than 20%.

JRF type 1, as employed herein for determination of fuel resistance, has the following composition (see AMS 2629, issued Jul. 1, 1989), section 3.1.1 et seq., available from SAE (Society of Automotive Engineers, Warrendale, Pa.):

Toluene 28±1% by volume

Cyclohexane (technical) 34±1% by volume

Isooctane 38±1% by volume

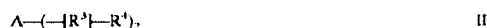
Tertiary dibutyl disulfide (doctor sweet) 1±0.005% by volume

Tertiary butyl mercaptan 0.015% ±0.0015 by weight of the other four components

Desirably, the inventive polythioethers have number average molecular weights ranging from about 500 to 20,000, preferably about 1,000 to 10,000, very preferably about 2,000 to 5,000.

Liquid polythioether polymers within the scope of the present invention can be difunctional, that is, linear polymers having two end groups, or polyfunctional, that is, branched polymers having three or more end groups. Depending on the relative amounts of dithiol(s) and divinyl ether(s) used to prepare the polymers, the polymers can have terminal thiol groups ( $\text{—SH}$ ) or terminal vinyl groups ( $\text{—CH=CH}_2$ ). Furthermore, the polymers can be uncapped, that is, include thiol or vinyl terminal groups that are not further reacted, or capped, that is, include thiol or vinyl groups that are further reacted with other compounds. Capping the polythioethers of the invention enables introduction of additional terminal functionalities, for example, hydroxyl or amine groups, to the inventive polymers, or in the alternative, introduction of end groups that resist further reaction, such as terminal alkyl groups.

A first preferred embodiment of the inventive polythioethers has the formula II



wherein

A denotes a structure having the formula I,

y is 0 or 1,

$\text{R}^3$  denotes a single bond when  $y=0$  and  $\text{—S—(CH}_2\text{)}_2\text{—}[\text{—O—R}^2\text{—}]_m\text{—O—}$  when  $y=1$ ,

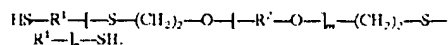
$\text{R}^4$  denotes  $\text{—SH}$  or  $\text{—S—(—CH}_2\text{—)}_2\text{—O—R}^5$  when  $y=0$  and  $\text{—CH}_2\text{=CH}_2$  or  $\text{—(CH}_2\text{—)}_2\text{—S—R}^5$  when  $y=1$ ,

$\text{R}^5$  denotes  $\text{C}_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $\text{—OH}$  or  $\text{—NHR}^7$  group, and

$\text{R}^7$  denotes H or a  $\text{C}_{1-6}$  n-alkyl group.

Thus, polythioethers of the formula II are linear, difunctional polymers which can be uncapped or capped. When  $y=0$ , the polymer includes terminal thiol groups or capped derivatives thereof. When  $y=1$ , the polymer includes terminal vinyl groups or capped derivatives thereof.

According to one preferred embodiment, the inventive polythioether is a difunctional thiol-terminated (uncapped) polythioether. That is, in formula II,  $y=0$  and  $\text{R}^4$  is  $\text{—SH}$ . Thus, the polythioether has the following structure:



The foregoing polymers are produced, for example, by reacting a divinyl ether or mixture thereof with an excess of a dithiol or mixture thereof, as discussed in detail below.

In a more particular preferred embodiment of the foregoing polythioether, when  $m=1$  and  $R^2=n$ -butyl in formula II,  $R^1$  is not ethyl or  $n$ -propyl. Also preferably, when  $m=1$ ,  $p=2$ ,  $q=2$ ,  $r=2$  and  $R^2=ethyl$ ,  $X$  is not O.

According to another preferred embodiment, the inventive polythioether is a capped polymer in which the foregoing terminal  $-SH$  groups are replaced by  $-S-(CH_2)_2-O-R^5$ . Such caps are produced by reaction of the terminal thiol group with a monovinyl ether, for example by including in the reaction mixture a capping agent or mixture thereof, as discussed in detail below.

In the foregoing,  $R^5$  denotes an unsubstituted or substituted alkyl group, preferably a  $C_{1-6}$   $n$ -alkyl group which is unsubstituted or substituted with at least one  $-OH$  or  $-NHR^7$  group, with  $R^7$  denoting H or  $C_{1-6}$   $n$ -alkyl. Exemplary useful  $R^5$  groups include alkyl groups, such as ethyl, propyl and butyl; hydroxyl-substituted groups such as 4-hydroxybutyl; amine-substituted groups such as 3-aminopropyl; etc.

Polythioethers according to the invention also include difunctional vinyl-terminated (uncapped) polythioethers. That is, in formula II,  $y=1$  and  $R^4$  is  $-CH=CH_2$ . These polymers are produced, for example, by reacting a dithiol or mixture thereof with an excess of a divinyl ether or mixture thereof, as discussed in detail below. Analogous capped polythioethers include terminal  $-(CH_2)_2-S-R^5$ .

The foregoing polythioethers are linear polymers having a functionality of 2 (considering alkyl and other non-reactive caps within this total). Polythioethers having higher functionality are also within the scope of the present invention. Such polymers are prepared, as discussed in detail below, by using a polyfunctionalizing agent. The term "polyfunctionalizing agent" as employed herein denotes a compound having more than two moieties that are reactive with terminal  $-SH$  and/or  $-CH=CH_2$  groups. The polyfunctionalizing agent preferably includes from 3 to 6 such moieties, and thus is denoted a " $z$ -valent" polyfunctionalizing agent, where  $z$  is the number (preferably from 3 to 6) of such moieties included in the agent, and hence the number of separate branches which the polyfunctional polythioether comprises. The polyfunctionalizing agent can be represented by the formula



where  $R^6$  denotes a moiety that is reactive with terminal  $-SH$  or  $-CH=CH_2$  and can be the same or different, and  $B$  is the  $z$ -valent residue of the polyfunctionalizing agent, i.e., the portion of the agent other than the reactive moieties  $R^6$ .

Polyfunctional polythioethers according to the present invention thus preferably have the formula III



wherein

$A$  denotes a structure having the formula 1,

$y$  is 0 or 1,

$R^3$  denotes a single bond when  $y=0$  and  $-S-(CH_2)_2-[O-R^2]_m-O-$  when  $y=1$ ,

$R^4$  denotes  $-SH$  or  $-S-(CH_2)_2-O-R^5$  when  $y=0$  and  $-CH=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,

$R^5$  denotes  $C_{1-6}$   $n$ -alkyl which is unsubstituted or substituted with at least one  $-OH$  or  $-NHR^7$  group,

$R^7$  denotes H or a  $C_{1-6}$   $n$ -alkyl group,

$z$  is an integer from 3 to 6, and

$B$  denotes a  $z$ -valent residue of a polyfunctionalizing agent.

As with the preceding difunctional embodiments, the foregoing polyfunctional polythioethers of the present invention can include terminal  $-SH$  or  $-CH=CH_2$  groups, or can be capped and thus include terminal  $-S-(CH_2)_2-O-R^5$  or  $-(CH_2)_2-S-R^5$  groups. Partially capped polyfunctional polymers, i.e., polymers in which some but not all of the branches are capped, are also within the scope of the present invention.

Specific polyfunctionalizing agents include trifunctionalizing agents, that is, compounds with  $z=3$ . Preferred trifunctionalizing agents include triallylcyanurate (TAC), which is reactive with compounds of the formula II ( $R^6=allyl$ ), and 1,2,3-propanetriethiol, which is reactive with compounds of the formula III ( $R^6=SH$ ).

Agents having mixed functionality, i.e., agents that include moieties (typically separate moieties) that react with both thiol and vinyl groups, can also be employed.

Other useful polyfunctionalizing agents include trimethylolpropane trivinyl ether, and the polythiols described in U.S. Pat. No. 4,366,307, U.S. Pat. No. 4,609,762 and U.S. Pat. No. 5,225,472, the disclosures of each of which are incorporated in their entireties herein by reference.

Polyfunctionalizing agents having more than three reactive moieties (i.e.,  $z>3$ ) afford "star" polythioethers and hyperbranched polythioethers. For example, two equivalents of TAC can be reacted with one mole of a dithiol to afford a material having an average functionality of 4. This material can then be reacted with a divinyl ether and a dithiol to yield a polymer, which can in turn be mixed with a trifunctionalizing agent to afford a polymer blend having an average functionality between 3 and 4.

Polythioethers as described above have a wide range of average functionality.

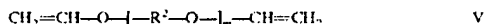
For example, trifunctionalizing agents afford average functionalities from about 2.05 to 3.0, preferably about 2.1 to 2.6. Wider ranges of average functionality can be achieved by using quaternary or higher polyfunctionalizing agents. Functionality will also be affected by factors such as stoichiometry, as is known to those skilled in the art.

Methods of making the foregoing polyfunctional polythioethers are discussed in detail below.

Polythioethers within the scope of the present invention are prepared by a number of methods. According to a first preferred method,  $(n+1)$  equivalents of a compound having the formula IV



or a mixture of at least two different compounds having the formula IV, are reacted with  $n$  equivalents of a compound having the formula V



or a mixture of at least two different compounds having the formula V, in the presence of a catalyst. In formulas IV and V above,  $R^1$ ,  $R^2$  and all indices are defined as in formula I. This method affords an uncapped, thiol-terminated difunctional polythioether.

The compounds of formula IV are dithiol compounds. Preferred dithiols include those compounds in which  $R^1$  is a divalent  $C_{2-6}$   $n$ -alkyl group, i.e., 1,2-ethanedithiol, 1,3-propanedithiol, 1,4-butanedithiol, 1,5-pentanedithiol or 1,6-hexanedithiol.

Additional preferred dithiols include those compounds in which  $R^1$  is a divalent  $C_{3-6}$  branched alkyl group, having one or more pendent groups which can be, for example, methyl or ethyl groups. Preferred compounds having branched alkyl  $R^1$  include 1,2-propanedithiol, 1,3-butanedithiol, 2,3-butanedithiol, 1,3-pentanedithiol, 1,3-dithio-3-methylbutane and 2,3-butanedithiol. Other useful dithiols include those in which  $R^1$  is a divalent  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group, for example, dipentenedithiol and ethylcyclohexyldithiol (ECHDT).

Further preferred dithiols include one or more heteroatom substituents in the carbon backbone, that is, dithiols in which  $X$  is a heteroatom such as O, S or another bivalent heteroatom radical; a secondary or tertiary amine group, i.e.,  $-NR^6-$ , where  $R^6$  is hydrogen or methyl; or another substituted trivalent heteroatom. In a preferred embodiment,  $X$  is O or S, and thus  $R^1$  is  $-[(CH_2)_p-O]_q-(CH_2)_r-$  or  $-[(CH_2)_p-S]_q-(CH_2)_r-$ . Preferably, the indices  $p$  and  $r$  are equal, and very preferably both have the value of 2. Particularly preferred exemplary dithiols of this type include dimercaptodithiylsulfide (DMDS) ( $p, r=2, q=1, X=S$ ); dimercaptodioxaoctane (DMDO) ( $p, q, r=2, X=O$ ); and 1,5-dithio-3-oxapentane. It is also possible to employ dithiols that include both heteroatom substituents in the carbon backbone and pendent alkyl, in particular methyl, groups. Such compounds include methyl-substituted DMDS, such as  $HS-CH_2CH(CH_3)-S-CH_2CH_2-SH$ ,  $H_2S-CH(CH_3)CH_2-S-CH_2CH_2-SH$  and dimethyl substituted DMDS such as  $H_2S-CH(CH_3)CH_2-S-CH(CH_3)CH_2-SH$  and  $HS-CH(CH_3)CH_2-S-CH_2CH(CH_3)-SH$ .

Two or more different dithiols of formula IV can also be employed if desired in preparing polythioethers according to the invention.

The compounds of formula V are divinyl ethers. Divinyl ether itself ( $m=0$ ) can be used. Preferred divinyl ethers include those compounds having at least one alkoxy group, more preferably from 1 to 4 alkoxy groups (i.e., those compounds in which  $m$  is an integer from 1 to 4). Very preferably,  $m$  is an integer from 2 to 4. It is also possible to employ commercially available divinyl ether mixtures in producing polythioethers according to the invention. Such mixtures are characterized by a non-integral average value for the number of alkoxy units per molecule. Thus,  $m$  in formula V can also take on non-integral, rational values between 0 and 10, preferably between 1 and 10, very preferably between 1 and 4, particularly between 2 and 4.

Exemplary divinyl ethers include those compounds in which  $R^2$  is  $C_{2-6}$  n-alkyl or  $C_{2-6}$  branched alkyl. Preferred divinyl ethers of this type include ethylene glycol divinyl ether (EG-DVE) ( $R^2$ =ethyl,  $m=1$ ); butanediol divinyl ether (BD-DVE) ( $R^2$ =butyl,  $m=1$ ); hexanediol divinyl ether (HD-DVE) ( $R^2$ =hexyl,  $m=1$ ); diethylene glycol divinyl ether (DEG-DVE) ( $R^2$ =ethyl,  $m=2$ ); triethylene glycol divinyl ether ( $R^2$ =ethyl,  $m=3$ ); and tetraethylene glycol divinyl ether ( $R^2$ =ethyl,  $m=4$ ). Useful divinyl ether blends include "PLURIOL®" type blends such as PLURIOL® E-200 divinyl ether (commercially available from BASF), for which  $R^2$ =ethyl and  $m=3.8$ , as well as "DPE" polymeric blends such as DPE-2 and DPE-3 (commercially available from International Specialty Products, Wayne, N.J.). Of these, DEG-DVE and PLURIOL® E-200 are particularly preferred.

Useful divinyl ethers in which  $R^2$  is  $C_{2-6}$  branched alkyl can be prepared by reacting a polyhydroxy compound with acetylene. Exemplary compounds of this type include compounds in which  $R^2$  is an alkyl-substituted methylene group such as  $-CH(CH_3)-$  or  $-CH_2CH(CH_3)-$ .

Other useful divinyl ethers include compounds in which  $R^2$  is polytetrahydrofuryl (poly-THF) or polyoxyalkylene, preferably having an average of about 3 monomer units.

Two or more compounds of the formula V can be used in the foregoing method. Thus in preferred embodiments of the invention, two compounds of formula IV and one compound of formula V, one compound of formula IV and two compounds of formula V, two compounds of formula IV and of formula V, and more than two compounds of one or both formulas, can be used to produce a variety of polythioethers according to the invention, and all such combinations of compounds are contemplated as being within the scope of the invention.

Although, as indicated above, compounds of the formulas IV and V which have pendent alkyl groups, for example pendent methyl groups, are useful according to the invention, it has surprisingly been discovered that compounds of the formulas IV and V which are free of pendent methyl or other alkyl groups also afford polythioethers that are liquid at room temperature and pressure.

The reaction between the compounds of formulas IV and V is preferably catalyzed by a free radical catalyst. Preferred free radical catalysts include azo compounds, for example azobisisobutyronitrile compounds such as azo(bis)isobutyronitrile (AIBN); organic peroxides such as benzoyl peroxide and *t*-butyl peroxide; and inorganic peroxides such as hydrogen peroxide. The reaction can also be effected by irradiation with ultraviolet light either with or without a cationic photoinitiating moiety. Ionic catalysis methods, using either inorganic or organic bases, e.g., triethylamine, also yield materials useful in the context of this invention.

Capped analogs to the foregoing polythioethers can be prepared by reacting ( $n+1$ ) equivalents of a compound having the formula IV or a mixture of at least two different compounds having the formula IV, ( $n$ ) equivalents of a compound having the formula V or a mixture of at least two different compounds having the formula V, and about 0.05 to about 2 equivalents of a compound having the formula VI



or a mixture of two different compounds having the formula VI, in the presence of an appropriate catalyst.

Compounds of the formula VI are monovinyl ethers, which react with terminal thiol groups to cap the polythioether polymer. Preferred monovinyl ethers of the formula VI include amino- and hydroxyalkyl vinyl ethers, such as 3-aminopropyl vinyl ether and 4-hydroxybutyl vinyl ether (butanediol monovinyl ether), as well as unsubstituted alkyl vinyl ethers such as ethyl vinyl ether. Use of 2 equivalents of compounds of the formula VI affords fully capped polymers, while use of lesser amounts results in partially capped polymers.

According to another preferred method, ( $n$ ) equivalents of a compound having the formula IV, or a mixture of at least two different compounds having the formula IV, are reacted with ( $n+1$ ) equivalents of a compound having the formula V, or a mixture of at least two different compounds having the formula V, again in the presence of an appropriate catalyst. This method affords an uncapped, vinyl-terminated difunctional polythioether.

Capped analogs to the foregoing vinyl-terminated polythioethers can be prepared by reacting ( $n+1$ ) equivalents of a compound having the formula V or a mixture of at least two different compounds having the formula V, ( $n$ ) equivalents of a compound having the formula IV or a mixture of at least two different compounds having the formula IV, and about 0.05 to about 2 equivalents of a compound having the formula VII

or a mixture of two different compounds having the formula VII, in the presence of an appropriate catalyst.

Compounds of the formula VII are monothiols, which can be unsubstituted or substituted with, e.g., hydroxyl or amino groups. Exemplary capping compounds of the formula VII include mercaptoalcohols such as 3-mercaptoopropanol, and mercaptoamines such as 4-mercaptobutylamine.

Polyfunctional analogs of the foregoing difunctional polythioethers are similarly prepared by combining one or more compounds of formula IV and one or more compounds of formula V, in appropriate amounts, with a polyfunctionalizing agent as described above, and reacting the mixture. Thus, according to one method for making polyfunctional polythioethers of the present invention, (n+1) equivalents of a compound or compounds having the formula IV, (n) equivalents of a compound or compounds having the formula V, and a z-valent polyfunctionalizing agent, are combined to form a reaction mixture. The mixture is then reacted in the presence of a suitable catalyst as described above to afford thiol-terminated polyfunctional polythioethers. Capped analogs of the foregoing polythioethers are prepared by inclusion in the starting reaction mixture of about 0.05 to about (z) equivalents one or more appropriate capping compounds VI. Use of (z) equivalents affords fully capped polyfunctional polymers, while use of lesser amounts again yields partially capped polymers.

Similarly, (n) equivalents of a compound or compounds having the formula IV, (n+1) equivalents of a compound or compounds having the formula V, and a z-valent polyfunctionalizing agent, are combined to form a reaction mixture and reacted as above to afford vinyl-terminated polyfunctional polythioethers. Capped analogs of the foregoing polythioethers are prepared by inclusion in the starting reaction mixture of one or more appropriate capping compounds VII.

The inventive polythioethers preferably are prepared by combining at least one compound of formula IV and at least one compound of formula V, optionally together with one or capping compounds VI and/or VII as appropriate, and/or a polyfunctionalizing agent, followed by addition of an appropriate catalyst, and carrying out the reaction at a temperature from about 30 to about 120° C. for a time from about 2 to about 24 hours. Very preferably the reaction is carried out at a temperature from about 70 to about 90° C. for a time from about 2 to about 6 hours.

Since the inventive reaction is an addition reaction, rather than a condensation reaction, the reaction typically proceeds substantially to completion, i.e., the inventive polythioethers are produced in yields of approximately 100%. No or substantially no undesirable by-products are produced. In particular, the reaction does not produce appreciable amounts of malodorous cyclic by-products such as are characteristic of known methods for producing polythioethers. Moreover, the polythioethers prepared according to the invention are substantially free of residual catalyst. As a result, no free catalyst is available to further react with the polythioether, in particular in the presence of water at room temperature, to degrade the polymer and produce malodorous cyclic compounds. Thus, the inventive polythioethers are characterized both by thermal stability and by low odor.

Polythioethers according to the invention are useful in applications such as coatings and sealant compositions, and preferably are formulated as polymerizable sealant compositions in applications where low temperature flexibility and fuel resistance are important. Such sealant compositions are useful, e.g., as aerospace sealants and linings for fuel tanks. A first preferred polymerizable composition thus includes at

least one polythioether as described herein; a curing agent or combination of curing agents; and a filler.

The polythioether or combination of polythioethers preferably is present in the polymerizable composition in an amount from about 30 wt % to about 90 wt %, more preferably about 40 to about 80 wt %, very preferably about 45 to about 75 wt %, with the wt % being calculated based on the weight of all non-volatile components of the composition. Preferably, the T<sub>g</sub> of the polythioether(s) used in the polymerizable composition is not higher than -55° C., more preferably not higher than -60° C.

Curing agents useful in polymerizable compositions of the invention include epoxy resins, for example, hydantoin diepoxide, diglycidyl ether of bisphenol-A epoxides, diglycidyl ether of bisphenol-F epoxides, Novolak type epoxides, and any of the epoxidized unsaturated and phenolic resins. Other useful curing agents include unsaturated compounds such as acrylic and methacrylic esters of commercially available polyols, unsaturated synthetic or naturally occurring resin compounds, TAC, and olefinic terminated derivatives of the compounds of the present invention. In addition, useful cures can be obtained through oxidative coupling of the thiol groups using organic and inorganic peroxides (e.g., MnO<sub>2</sub>) known to those skilled in the art. Selection of the particular curing agent may affect the T<sub>g</sub> of the cured composition. For example, curing agents that have a T<sub>g</sub> significantly lower than the T<sub>g</sub> of the polythioether may lower the T<sub>g</sub> of the cured composition.

Depending on the nature of the polythioether(s) used in the composition, the composition will contain about 90% to about 150% of the stoichiometric amount, preferably about 95 to about 125%, of the selected curing agent(s).

Fillers useful in the polymerizable compositions of the invention include those commonly used in the art, such as carbon black and calcium carbonate (CaCO<sub>3</sub>). Preferably, the compositions include about 5 to about 60 wt % of the selected filler or combination of fillers, very preferably about 10 to 50 wt %.

The polythioethers, curing agents and fillers employed in polymerizable compositions of the invention, as well as optional additives as described below, should be selected so as to be compatible with each other. Selection of compatible ingredients for the inventive compositions can readily be performed by those skilled in the art without recourse to undue experimentation.

The foregoing polymerizable compositions preferably are curable at a minimum temperature of about 0° C. (i.e., at a temperature of about 0° C. or higher), more preferably about -10° C., very preferably about -20° C., and have a T<sub>g</sub> when cured not higher than about -55° C., more preferably not higher than -60° C., very preferably not higher than -65° C. When cured, the polymerizable compositions preferably have a % volume swell not greater than 25%, more preferably not greater than 20%, after immersion for one week at 60° C. (140° F.) and ambient pressure in jet reference fluid (JRF) type 1.

In addition to the foregoing ingredients, polymerizable compositions of the invention can optionally include one or more of the following: pigments; thixotropes; accelerators; retardants; adhesion promoters; and masking agents.

Useful pigments include those conventional in the art, such as carbon black and metal oxides. Pigments preferably are present in an amount from about 0.1 to about 10 wt %.

Thixotropes, for example silica, are preferably used in an amount from about 0.1 to about 5 wt %.

Accelerators known to the art, such as amines, preferably are present in an amount from about 0.1 to about 5 wt %.

Two such useful accelerators are 1,4-diaza-bicyclo[2.2.2]octane (DABCO®, commercially available from Air Products, Chemical Additives Division, Allentown, Pa.) and DMP-30® (an accelerator composition including 2,4,6-tri(dimethylaminomethyl)phenol, commercially available from Rohm and Haas, Philadelphia, Pa.).

Retardants, such as stearic acid, likewise preferably are used in an amount from about 0.1 to about 5 wt %. Adhesion promoters, which can be, for example, conventional phenolics or silanes, if employed are preferably present in amount from about 0.1 to about 5 wt %. Masking agents, such as pine fragrance or other scents, which are useful in covering any low level odor of the composition, are preferably present in an amount from about 0.1 to about 1 wt %.

An additional advantage of sealant compositions according to the invention is their improved curing behavior. The extent of cure of a sealant composition as a function of time is often difficult to measure directly, but can be estimated by determining the extrusion rate of the composition as a function of time. The extrusion rate is the rate at which a mixed sealant composition, i.e., a sealant composition together with an accelerator system, is extruded from an applicator device. Since the sealant composition is mixed with the accelerator system, curing begins, and the extrusion rate changes with time. The extrusion rate thus is inversely related to the extent of cure. That is, when the extent of cure is low, the viscosity of the mixed sealant composition is low and thus the extrusion rate is high. When the reaction approaches completion, the viscosity becomes very high, and the extrusion rate thus becomes low.

With reference to FIG. 1, the viscosity of some known types sealant compositions remains low for an extended time, because the compositions are slow to cure. Such compositions have extrusion curves qualitatively similar to curve A. Other known types of sealant composition cure very quickly, and thus their viscosity rapidly increases. Consequently, the extrusion rate rapidly decreases, as shown in curve B. Desirably, a mixed sealant composition should have a low viscosity, and thus a high extrusion rate, for a length of time sufficient to allow even application of the sealant composition to the area requiring sealing, but then should cure rapidly after application, i.e., their extrusion rate should quickly decrease. Sealant compositions according to the present invention are characterized by this desirable extrusion curve, as illustrated qualitatively in curve C.

Sealant compositions according to the present invention can have, depending on the particular formulation, initial extrusion rates as high as 500 g/min or higher, together with low extrusion rates on the order of about 5 to 10 g/min or less after curing times on the order of one hour.

As shown in FIG. 2, the initial extrusion rate of a polymer of the present invention (Example 1, below, cured with an epoxy curing agent as described below) is about 550 g/min, then falls rapidly to about 20 g/min after 70 minutes. In comparison, a known polysulfide (cured with  $MnO_2$ ) has an initial extrusion rate of about 90 g/min, which slowly falls to about 20 g/min after 70 minutes.

A second preferred polymerizable composition combines one or more plasticizers with the polythioether(s), curing agent(s) and filler(s) described above. Use of a plasticizer allows the polymerizable composition to include polythioethers which have higher  $T_g$  than would ordinarily be useful in an aerospace sealant. That is, use of a plasticizer effectively reduces the  $T_g$  of the composition, and thus increases the low-temperature flexibility of the cured polymerizable composition beyond that which would be expected on the basis of the  $T_g$  of the polythioethers alone.

Plasticizers that are useful in polymerizable compositions of the invention include phthalate esters, chlorinated paraffins, hydrogenated terphenyls, etc. The plasticizer or combination of plasticizers preferably constitute 1 to about 40 wt %, more preferably 1 to about 10 wt % of the composition.

Depending on the nature and amount of the plasticizer(s) used in the composition, polythioethers of the invention which have  $T_g$  values up to about  $-50^\circ\text{C}$ ., preferably up to about  $-55^\circ\text{C}$ ., can be used.

The foregoing polymerizable compositions also preferably are curable at a minimum temperature of about  $0^\circ\text{C}$ ., more preferably about  $-10^\circ\text{C}$ ., very preferably about  $-20^\circ\text{C}$ .

The present invention is illustrated in more detail by means of the following non-limiting examples.

In examples 1-8, liquid polythioethers were prepared by stirring together one or more dithiols with one or more divinyl ethers and a trifunctionalizing agent. The reaction mixture was then heated and a free radical catalyst was added. All reactions proceeded substantially to completion (approximately 100% yield).

#### Example 1

In a 2 L flask, 524.8 g (3.32 mol) of diethylene glycol divinyl ether (DEG-DVE) and 706.7 g (3.87 mol) of dimercaptodioxaoctane (DMDO) were mixed with 19.7 g (0.08 mol) of triallylcyanurate (TAC) and heated to  $77^\circ\text{C}$ . To the heated reaction mixture was added 4.6 g (0.024 mol) of an azobisisnitrile free radical catalyst (VAZO® 67 [2,2'-azobis(2-methylbutyronitrile), commercially available from DuPont]). The reaction proceeded substantially to completion after 2 hours to afford 1250 g (0.39 mol, yield 100%) of a liquid polythioether resin having a  $T_g$  of  $-68^\circ\text{C}$ . and a viscosity of 65 poise. The resin was faintly yellow and had low odor.

#### Example 2

In a 1 L flask, 404.4 g (1.60 mol) of PLURIOL® E-200 divinyl ether and 355.88 g (1.94 mol) of DMDO were mixed with 12.1 g (0.049 mol) of TAC and reacted as in Example 1. The reaction proceeded substantially to completion after 5 hours to afford 772 g (0.024 mol, yield 100%) of a resin having a  $T_g$  of  $-66^\circ\text{C}$ . and a viscosity of 48 poise. The resin was yellow and had low odor.

#### Example 3

In a 100 mL flask, 33.2 g (0.21 mol) of DEG-DVE and 26.48 g (0.244 mol) of 1,2-propanedithiol were mixed with 0.75 g (0.003 mol) of TAC and heated to  $71^\circ\text{C}$ . To the heated reaction mixture was added 0.15 g (0.8 mmol) of VAZO® 67. The reaction proceeded substantially to completion after 7 hours to afford 60 g (0.03 mol, yield 100%) of a resin having a  $T_g$  of  $-61^\circ\text{C}$ . and a viscosity of 22 poise. The resin had a noticeable PDT odor.

#### Example 4

In a 100 mL flask, 33.3 g (0.136 mol) of tripropylene glycol divinyl ether (DPE-3) and 27.0 g (0.170 mol) of dimercaptodimethylsulfide (DMDS) were mixed with 0.69 g (0.003 mol) of TAC and heated to  $77^\circ\text{C}$ . To the heated reaction mixture was added 0.15 g (0.8 mmol) of VAZO® 67. The reaction proceeded substantially to completion after 6 hours to afford 61 g (0.028 mol, yield 100%) of a resin having a  $T_g$  of  $-63^\circ\text{C}$ . and a viscosity of 26 poise.

## Example 5

In a 250 ml. flask, 113.01 g (0.447 mol) of PLURIOL® E-200 divinyl ether and 91.43 g (0.498 mol) of DMDO were mixed with 1.83 g (0.013 mol) of 1,2,3-propanetrithiol (PTT) and allowed to react exothermically for 72 hours. The mixture was then heated to 80° C. To the heated reaction mixture was added 0.2 g (1 mmol) of VAZO® 67. The reaction mixture was maintained at 80° C., and the reaction proceeded substantially to completion after 3 hours to afford 200 g (0.06 mol, yield 100%) of a resin having a  $T_g$  of -66° C. and a viscosity of 55 poise.

## Example 6

In a small jar, 14.0 g (0.055 mol) of PLURIOL® E-200 divinyl ether, 6.16 g (0.336 mol) of DMDO and 5.38 g (0.336 mol) of DMDS were mixed with 0.42 g (0.017 mol) of TAC (briefly heated to melt the TAC) and heated to 82° C. To the heated reaction mixture was added 0.2 g (0.001 mol) of VAZO® 67. The reaction proceeded substantially to completion after 18 hours to afford 26 g (8.4 mmol, yield 100%) of a resin having a  $T_g$  of -63° C. and a viscosity of 80 poise.

## Example 7

In a small jar, 13.55 g (0.054 mol) of PLURIOL® E-200 divinyl ether, 10.44 g (0.057 mol) of DMDO and 1.44 g (8.1 mmol) of ethylcyclohexanedithiol (ECHDT) were mixed with 0.40 g (1.6 mmol) of TAC (heated briefly to melt the TAC) and heated to 82° C. To the heated reaction mixture was added 0.2 g (0.001 mol) of VAZO® 67. The reaction proceeded substantially to completion after 5 hours to afford 26 g (8.1 mmol, yield 100%) of a resin having a  $T_g$  of -66° C. and a viscosity of 58 poise.

## Example 8

In a small glass jar, 9.11 g (0.036 mol) of PLURIOL® E-200 divinyl ether, 5.71 g (0.031 mol) of DMDO, 1.52 g (7.8 mmol) of ECHDT, 5.08 g (0.031 mol) of DMDS and 4.11 g (0.024 mol) of hexanediol divinyl ether (HD-DVE) were mixed with 0.39 g (1.6 mmol) of TAC (heated briefly to dissolve the TAC) and heated to 82° C. To the heated reaction mixture was added 0.6 g (3.1 mmol) of VAZO® 67. The reaction proceeded substantially to completion after about 45 hours to afford 2.6 g (7.8 mmol, yield 100%) of a resin having a  $T_g$  of -66° C. and a viscosity of 304 poise. The resin had a cloudy appearance.

Each of the foregoing resins was evaluated for odor. The following scale was employed: 3: strong, offensive odor; 2: moderate odor; 1: slight odor; 0: substantially odorless.

The polymer described in Example 3 of U.S. Pat. No. 4,366,307 was used as a control. This polymer (the "control polymer") had an odor of 3.

Results were as follows:

Polymer	Odor	Polymer	Odor
1	1	5	1
2	1	6	1
3	3	7	1
4	1	8	2

All of the liquid polythioethers thus had little or moderate odor except polymer 3, which had a strong odor.

The resins prepared in Examples 1-8 were then cured. Curing was carried out using the uncompound resins with a curing agent and DABCO accelerator. The curing agent had the following composition:

epoxy novolak (equivalent weight 175.5)	22 wt %
hydantoin epoxy (equivalent weight 132)	34 wt %
calcium carbonate	34 wt %
carbon black	5 wt %
silane adhesive promoter	5 wt %

The cured resins were evaluated for odor according to the procedure set forth above. The  $T_g$  and the percent weight gain after immersion in JRF type 1 for one week at room temperature and pressure were also measured for each of the cured resins. The volume swell and weight gain percentages were determined for each cured material as follows:

$w_1$  = initial weight in air

$w_2$  = initial weight in  $H_2O$

$w_3$  = final weight in air

$w_4$  = final weight in  $H_2O$

% volume swell =  $100 \times [(w_2 + w_3) - (w_1 + w_4)] / (w_1 - w_2)$

% weight gain =  $100 \times (w_3 - w_1) / w_1$

The results are given in Table 1:

TABLE 1

Cured Resin	1	2	3	4	5	6	7	8
Odor	0	0	0	0	0	0	0	0
$T_g$ (°C.)	-59	-61	-61	-63	-62	-56	-59	-58
% fuel swell	19	22	—	—	23	19	24	27
% wt gain	14	15	15	23	15	15	19	20

In comparison, the control polymer had an odor of 1-2 when cured.

## Example 9

Polythioethers having a number average molecular weight of 2100 and an average functionality  $F$  of 2.1 were prepared by combining a divinyl ether with a dithiol as shown in Table 2 and reacting the materials as described herein. The uncompound polythioethers were then cured using 15 g of the curing agent described above and 0.30 g of DABCO. For each polythioether so prepared, the following quantities were measured: viscosity (uncured material, poise p); Shore A hardness (cured material, Rex durometer value); % weight gain (cured material) after one week at 140° F. (60° C.) and atmospheric pressure in JRF type 1; and  $T_g$  (uncured material, ° C.). Results were as follows:

TABLE 2

dithiol divinyl ether	ECHDT	DMDS	DMDO	HDT <sup>a</sup>
DEG-DVE	145 p	(solid)	27 p	24 p
	44 Rex	94 Rex	25 Rex	25 Rex
	27%	3%	14%	29%
	-53	-63	-69	-77
PLURIOL® *	77 p	41 p	59 p	25 p
	43 Rex	47 Rex	27 Rex	23 Rex
	27%	11%	18%	30%
	-57	-61	-67	-76
BD-DVE <sup>b</sup>	185 p	(solid)	(solid)	(solid)
	42 Rex	—	20 Rex	22 Rex
	44%	—	21%	44%
	-59	—	-79	-85
HD-DVE	155 p	(solid)	(solid)	(soft solid)
	50 Rex	—	14 Rex	29 Rex

TABLE 2-continued

dithiol divinyl ether	ECHDT	DMDS	DMDO	HDT <sup>d</sup>
	57%	—	27%	68%
	-60	-63	-78	-86
Poly-THF <sup>c</sup>	91 p	(solid)	27 p	—
	30 Rex	75 Rex	17 Rex	—
	64%	29%	37%	—
	-69	-79	-79	—

<sup>a</sup>PLURIOL® E-200 divinyl ether<sup>b</sup>Butanediol divinyl ether<sup>c</sup>polytetrahydrofuran divinyl ether<sup>d</sup>Hexanedithiol

From the foregoing table it is apparent that the following combinations of divinyl ether and dithiol afford liquid polythioethers having unexpectedly superior fuel resistance and low temperature flexibility when cured: PLURIOL® E-200/DMDO; and DEG-DVE/DMDO. Other potentially useful combinations include DEG-DVE/ECHDT; DEG-DVE/HDT; PLURIOL® E-200/ECHDT; PLURIOL® E-200/HDT; and poly-THF/DMDO. PLURIOL® E-200/DMDS also has excellent fuel resistance and low temperature flexibility when cured, but the uncompounded material does not remain in a liquid state for an extended period of time.

## Example 10

Addition of DMDS to PLURIOL®/DMDO  
Polymers

Four liquid polythiols were prepared as described herein. The polymers had the following compositions (listed values are molar equivalents):

	1	2	3	4
PLURIOL®	6.6	6.6	6.6	6.6
E-200	8	6	4.5	4
DMDO	0	2	3.5	4
DMDS				

Each uncompounded polymer was cured as in Example 9 (1.5 g of the curing agent composition and 0.30 g of DABCO), with the addition of 0.2 molar equivalents of TAC to afford polymers having a number average molecular weight of about 3000 and a functionality F of 2.2. For each polymer, the following properties were measured:  $T_g$  (resin, °C.);  $T_g$  (cured, °C.); viscosity (p); % swell in JRF type 1; % weight gain in JRF type 1; and % weight gain in water. Results are given in Table 3.

TABLE 3

	1	2	3	4
$T_g$ (resin)	-67	-66	-64	-63
(cured)	-59	-58	-56	-56
Viscosity	59	53	62	80
JRF				
% Swell	24	21	21	20
% Wt Gain	18	15	16	16
H <sub>2</sub> O	11.8	11.5	7.4	7.5
% Wt Gain				

All of the foregoing polymers displayed excellent fuel resistance. Polymers 1 and 2 in particular also displayed excellent low temperature flexibility.

## Example 11

Addition of ECHDT to PLURIOL®/DMDO  
Polymers

Four liquid polythiols were prepared as described herein. The polymers had the following compositions (listed values are molar equivalents):

	1	2	3	4
PLURIOL®	6.6	6.6	6.6	6.6
E-200	8	7	6	5
DMDO	0	1	2	3
ECHDT				

Each uncompounded polymer was cured as in Example 10 to afford polymers having a number average molecular weight of about 3000 and a functionality F of 2.2. For each polymer, the following properties were measured:  $T_g$  (resin, °C.);  $T_g$  (cured, °C.); viscosity (p); % swell in JRF type 1; % weight gain in JRF type 1; and % weight gain in water. Results are given in Table 4.

TABLE 4

	1	2	3	4
$T_g$ (resin)	-67	-66	-65	-64
(cured)	-59	-59	-58	-56
Viscosity	59	36	44	50
JRF type 1				
% Swell	24	25	28	29
% Wt Gain	18	18	19	19
H <sub>2</sub> O	11.8	10.8	8.3	7.8
% Wt Gain				

All of the foregoing polymers displayed good fuel resistance and low temperature flexibility.

## Example 12

In a 250 mL 3-neck flask equipped with a stirrer, thermometer and condenser, 87.7 g (0.554 mol) of DEG-DVE and 112.3 g (0.616 mol) of DMDO are mixed and heated to 77° C. (about 170° F.). To the mixture is added 0.8 g (4.2 mmol) of VAZO® 67 catalyst. The reaction mixture is reacted at 82° C. (about 180° F.) for about 6 hours to afford 200 g (0.06 mol, yield 100%) of a low viscosity liquid polythioether resin having a thiol equivalent of 1625 and a functionality F of 2.0.

## Example 13

In a 250 mL 3-neck flask equipped with a stirrer, thermometer and condenser, 26.7 g (0.107 mol) of TAC, 56.4 g (0.357 mol) of DEG-DVE and 117.0 g (0.642 mol) of DMDO are mixed and heated to 77° C. (about 170° F.). To the mixture is added 0.8 g (4.2 mmol) of VAZO 67 catalyst. The reaction mixture is reacted at 82° C. (about 180° F.) for about 6 hours to afford 200 g (0.07 mol, yield 100%) of a high viscosity liquid polythioether resin having an equivalent of 800 and a functionality F of about 3.5.

## Example 14

## Sealant Composition

A sealant composition including the DMDO/DEG-DVE polythioether polymer of Example 1 was compounded as follows (amounts in parts by weight):

DMDO/DEG-DVE Polythioether	100
Calcium carbonate	60
Magnesium oxide	1
Phenolic resin	1
DMP-30	1
Isopropyl alcohol	3

The compounded polymer was mixed intimately with the epoxy resin curing agent of Examples 9-11 above, in the weight ratio of 10:1 and cured at ambient temperature and humidity. The following physical properties were obtained for the cured composition:

Cure hardness at 25° C.	60 Shore A
Tensile strength at break	550 psi
Elongation at break	600%
Notched tear strength	100 p/i
Low-temperature flexibility (AMS 3267 § 4.5.4.7)	passed

#### Example 15

##### Sealant Composition

A sealant composition including the ECHDT/DEG-DVE polythioether polymer of Example 9 was compounded as follows (amounts in parts by weight):

ECHDT/DEG-DVE Polythioether	100
Calcium carbonate	54
Hydrated aluminum oxide	20
Magnesium oxide	1
Phenolic resin	1
Hydrogenated terphenyl plasticizer	6
DMP-30	1
Isopropyl alcohol	3

The compounded polymer was mixed intimately with an epoxy resin curing agent in the weight ratio of 10:1 and cured at ambient temperature and humidity. The following physical properties were obtained for the cured composition:

Cure hardness at 25° C.	72 Shore A
Tensile strength at break	550 psi
Elongation at break	450%
Notched tear strength	85 p/i
Low-temperature flexibility	passed

#### Example 16

##### OH-Terminated Capped Polythioether

In a 500 ml flask, 275.9 g (1.09 mol) PLURIOI.® E-200 divinyl ether, 174.7 g (0.95 mol) DMDO, 28.7 g (0.30 mol) 3-mercaptopropanol and 1.83 g (7.3 mmol) TAC were mixed. The mixture was heated to 70° C., and 2.3 g (12 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85-90° C. for 4 hours to afford 480 g (0.15 mol, yield 100%) of a polymer having an equivalent weight of 1670 (number average molecular weight=3200, functionality F=2.05).

#### Example 17

##### OH-Terminated Capped Polythioether

In a 250 ml flask, 104.72 g (0.57 mol) DMDO, 80.73 g (0.51 mol) DEG-DVE and 14.96 g (0.13 mol) butanediol

monovinyl ether were mixed and heated to 75° C. To the heated mixture 0.60 g (3 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 75-85° C. for 6 hours to afford 200 g (0.064 mol, yield 100%) of a clear, nearly colorless polymer with very low odor and a viscosity of 79 poise at 20° C. The equivalent weight was 1570 (number average molecular weight=3200, functionality F=2.00).

#### Example 18

##### Vinyl-Terminated Polythioether

In a 250 ml flask, 97.63 g (0.53 mol) DMDO, 97.66 g (0.62 mol) DEG-DVE and 5.31 g (0.21 mol) TAC were mixed and heated to 70° C. To the heated mixture 0.80 g (4 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85-90° C. for 4 hours to afford 200 g (0.11 mol, yield 100%) of a low-odor polymer having a  $T_g$  of -68° C. and a viscosity of 25 poise at 20° C. The equivalent weight was 1570 (number average molecular weight=1900, functionality F=2.2).

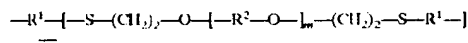
#### Example 19

##### Ethyl Vinyl Ether-Terminated Polythioether

In a 100 ml flask, 43.05 g (0.24 mol) DMDO, 34.22 g (0.22 mol) DEG-DVE and 2.84 g (0.04 mol) ethyl vinyl ether were mixed and heated to 80° C. To the heated mixture 0.28 g (1.5 mmol) VAZO® 67 were added slowly. The reaction mixture was stirred and heated at 85° C. for 6 hours to afford 80 g (0.02 mol, yield 100%) of a polymer having a  $T_g$  of -67° C. and a viscosity of 64 poise at 20° C. (number average molecular weight=4100, functionality F=2.0).

What is claimed is:

1. A polythioether comprising a structure having the formula I



wherein

$R^1$  denotes a divalent  $C_{2-6}$  n-alkyl,  $C_{3-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$ , or  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$  in which at least one  $\text{---CH}_2\text{---}$  unit is substituted with a methyl group,

$R^2$  denotes methylene, a divalent  $C_{2-6}$  n-alkyl,  $C_{2-6}$  branched alkyl,  $C_{6-8}$  cycloalkyl or  $C_{6-10}$  alkylcycloalkyl group,  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$  or  $\text{---}[(\text{---CH}_2\text{---})_p\text{---X---}]_q\text{---}(\text{---CH}_2\text{---})_r\text{---}$  in which at least one  $\text{---CH}_2\text{---}$  unit is substituted with a methyl group,

X is one selected from the group consisting of O, S and  $\text{---NR}^6\text{---}$ ,

$R^6$  denotes H or methyl,

m is a rational number from 0 to 10,

n is an integer from 1 to 60,

p is an integer from 2 to 6,

q is an integer from 1 to 5, and

r is an integer from 2 to 10,

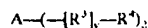
said polythioether being a liquid at room temperature and pressure.

2. The polythioether of claim 1 which has a glass transition temperature  $T_g$  not higher than -50° C.

3. The polythioether of claim 1 which, when cured, has a % volume swell not greater than 25% after immersion for one week in JRF type 1 at 60° C. and ambient pressure.

## 21

4. The polythioether of claim 1 which has a number average molecular weight between about 500 and 20,000.  
 5. The polythioether of claim 1 having the formula II



wherein

- A denotes a structure having the formula I,  
 y is 0 or 1,  
 $R^3$  denotes a single bond when  $y=0$  and  $-S-(CH_2)_2-$  10  
 $[-O-R^2-]_m-O-$  when  $y=1$ ,  
 $R^4$  denotes  $-SH$  or  $-S-(CH_2)_2-O-R^5$  when  $y=0$  and  $-CH_2=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,  
 $R^5$  denotes  $C_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $-OH$  or  $-NHR^7$  group, and 15  
 $R^7$  denotes H or a  $C_{1-6}$  n-alkyl group.  
 6. The polythioether of claim 5 wherein  $y=0$ .  
 7. The polythioether of claim 6 wherein  $R^4$  is  $-SH$ . 20  
 8. The polythioether of claim 7 wherein (i) when  $m=1$  and  $R^2=n$ -butyl,  $R^1$  is not ethyl or n-propyl, and (ii) when  $m=1$ ,  $p=2$ ,  $q=2$ ,  $r=2$  and  $R^2=ethyl$ , X is not O.  
 9. The polythioether of claim 6 wherein  $R^4$  is  $-S-(CH_2)_2-O-R^5$ . 25  
 10. The polythioether of claim 9 wherein  $R^5$  is  $n-C_2H_5$ ,  $n-C_4H_9-OH$  or  $n-C_3H_7-NH_2$ .  
 11. The polythioether of claim 5 wherein  $y=1$ .  
 12. The polythioether of claim 11 wherein  $R^4$  is  $-CH=CH_2$ . 30  
 13. The polythioether of claim 11 wherein  $R^4$  is  $-(CH_2)_2-S-R^5$ .  
 14. The polythioether of claim 13 wherein  $R^5$  is  $n-C_3H_7-OH$ .

## 22

15. The polythioether of claim 1 having the formula III



II 5 wherein

- A denotes a structure having the formula I,  
 y is 0 or 1,  
 $R^3$  denotes a single bond when  $y=0$  and  $-S-(CH_2)_2-$   
 $[-O-R^2-]_m-O-$  when  $y=1$ ,  
 $R^4$  denotes  $-SH$  or  $-S-(CH_2)_2-O-R^5$  when  $y=0$  and  $-CH_2=CH_2$  or  $-(CH_2)_2-S-R^5$  when  $y=1$ ,  
 $R^5$  denotes  $C_{1-6}$  n-alkyl which is unsubstituted or substituted with at least one  $-OH$  or  $-NHR^7$  group, 15  
 $R^7$  denotes H or a  $C_{1-6}$  n-alkyl group,  
 z is an integer from 3 to 6, and  
 B denotes a z-valent residue of a polyfunctionalizing agent.  
 16. The polythioether of claim 15 wherein  $z=3$ .  
 17. The polythioether of claim 16 which has an average functionality from about 2.05 to 3.00.  
 18. The polythioether of claim 15 wherein  $y=0$ .  
 19. The polythioether of claim 18 wherein  $R^4$  is  $-SH$ .  
 20. The polythioether of claim 18 wherein  $R^4$  is  $-S-(CH_2)_2-O-R^5$ .  
 21. The polythioether of claim 15 wherein  $y=1$ .  
 22. The polythioether of claim 21 wherein  $R^4$  is  $-CH=CH_2$ . 30  
 23. The polythioether of claim 21 wherein  $R^4$  is  $-(CH_2)_2-S-R^5$ .

\* \* \* \* \*

PATENT  
58179-5005

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re patent application of:  
Jonathan Doherty ZOOK, et al.

Serial No. 09/318,500

Filed: May 25, 1999

For: COMPOSITION AND METHOD  
FOR PRODUCING FUEL  
RESISTANT LIQUID  
POLYTHIOETHER POLYMERS  
WITH GOOD LOW  
TEMPERATURE FLEXIBILITY

Group Art Unit: 1714

Examiner:

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July 16, 1999

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re patent application of:  
Jonathan Doherty ZOOK, et al.

Serial No. 09/318,500

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For: COMPOSITION AND METHOD  
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RESISTANT LIQUID  
POLYTHIOETHER POLYMERS  
WITH GOOD LOW  
TEMPERATURE FLEXIBILITY

Group Art Unit: 1714

Examiner:

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Dear Sir:

In response to the Notice to File Missing Parts dated June 16,  
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1. a copy of the "Notice to File Missing Parts;"
2. a check for \$1376 to cover the statutory filing fee and surcharge fee.


Serial No. 09/318,500

The Commissioner is authorized to charge any additional amounts due in connection with this matter to our Deposit Account No. 10-0440. A duplicate copy of this letter is enclosed.

Respectfully submitted,

7/16/99

Date

  
\_\_\_\_\_  
DAVID J. MEYER  
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09/318, 500  
58179-5005

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APPLICATION NUMBER	FILING/RECEIPT DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NO./TITLE
09/318,500	05/25/99	ZOOK	J 53179-5002
JEFFER MANGELS BUTLER & MARMARO LLP TENTH FLOOR 2121 AVENUE OF THE STARS LOS ANGELES CA 90067			NOT ASSIGNED
			1714

DATE MAILED:

06/16/99

**NOTICE TO FILE MISSING PARTS OF APPLICATION**  
**Filing Date Granted**

An Application Number and Filing Date have been assigned to this application. The items indicated below, however, are missing. Applicant is given TWO MONTHS FROM THE DATE OF THIS NOTICE within which to file all required items and pay any fees required below to avoid abandonment. Extensions of time may be obtained by filing a petition accompanied by the extension fee under the provisions of 37 CFR 1.136(a). If any of items 1 or 3 through 5 are indicated as missing, the SURCHARGE set forth in 37 CFR 1.16(e) of ☐ \$65.00 for a small entity in compliance with 37 CFR 1.27, or ☒ \$130.00 for a non-small entity, must also be timely submitted in reply to this NOTICE to avoid abandonment.

If all required items on this form are filed within the period set above, the total amount owed by applicant as a

☐ small entity (statement filed) ☒ non-small entity is \$ 1310.

☒ 1. The statutory basic filing fee is:

- ☒ missing.  
☐ insufficient.

Applicant must submit \$ 760 to complete the basic filing fee and/or file a small entity statement claiming such status (37 CFR 1.27).

☒ 2. The following additional claims fees are due:

\$ 486 for 27 total claims over 20.

\$ \_\_\_\_\_ for \_\_\_\_\_ independent claims over 3.

\$ \_\_\_\_\_ for multiple dependent claim surcharge.

Applicant must either submit the additional claim fees or cancel additional claims for which fees are due.

☐ 3. The oath or declaration:

- ☐ is missing or unsigned.  
☐ does not cover the newly submitted items.

An oath or declaration in compliance with 37 CFR 1.63, including residence information and identifying the application by the above Application Number and Filing Date is required.

☐ 4. The signature(s) to the oath or declaration is/are by a person other than inventor or person qualified under 37 CFR 1.42, 1.43 or 1.47.

A properly signed oath or declaration in compliance with 37 CFR 1.63, identifying the application by the above Application Number and Filing Date, is required.

☐ 5. The signature of the following joint inventor(s) is missing from the oath or declaration:

An oath or declaration in compliance with 37 CFR 1.63 listing the names of all inventors and signed by the omitted inventor(s), identifying this application by the above Application Number and Filing Date, is required.

☐ 6. A \$50.00 processing fee is required since your check was returned without payment (37 CFR 1.21(m)).

☐ 7. Your filing receipt was mailed in error because your check was returned without payment.

☐ 8. The application was filed in a language other than English.

Applicant must file a verified English translation of the application, the \$130.00 set forth in 37 CFR 1.17(k), unless previously submitted, and a statement that the translation is accurate (37 CFR 1.52(d)).

☐ 9. OTHER:

Direct the reply and any questions about this notice to "Attention: Box Missing Parts."

**A copy of this notice MUST be returned with the reply.**

Customer Service Center  
Initial Patent Examination Division (703) 308-1202

## DECLARATION FOR PATENT APPLICATION

Docket Number (Optional)  
OC-573A1

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

**SEALANTS AND POTTING FORMULATIONS INCLUDING MERCAPTO-TERMINATED  
POLYMERS PRODUCED BY THE REACTION OF A POLYTHIOL AND POLYVINYL ETHER MONOMER,**

the specification of which is attached hereto unless the following box is checked:

☐ was filed on \_\_\_\_\_ as United States Application Number or PCT International Application Number  
and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Claimed

☐ Yes ☐ No

(Number)	(Country)	(Day/Month/Year Filed)
(Number)	(Country)	(Day/Month/Year Filed)

☐ Yes ☐ No

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

60/182,396	February 14, 2000
(Application Number)	(Filing Date)
60/215,548	June 30, 2000
(Application Number)	(Filing Date)

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

09/318,500	May 25, 1999	Pending
(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)
08/928,972	September 12, 1997	Pending
(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)
08/802,130	February 19, 1997	Patented
(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

William J. Uhl, Reg. No. 25,567  
Linda Pingitore, Reg. No. 30,414  
Deborah M. Altman, Reg. No. 42,259Jacques B. Miles, Reg. No. 42,888  
William E. Kuss, Reg. No. 41,919

Address all telephone calls to WILLIAM J. UHL at telephone number (412) 434-2881.

Address all correspondence to: PPG INDUSTRIES, INC.  
Intellectual Property Department  
One PPG Place  
Pittsburgh, Pennsylvania 15272

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor (given name, family name): Jonathan D. Zook

Inventor's signature \_\_\_\_\_

Date,  
City

Residence: Santa Clarita, California, United States of America

Post Office Address: 20303 High Point Place

Santa Clarita, California 91351

ZOOK EXHIBIT 1035

Zook v. Zook

Interference No. 105,555 (MPT)

☒ Additional inventors are being named on separately numbered sheets attached hereto.

## DECLARATION FOR PATENT APPLICATION

Docket Number (Optional)  
OC-573A1

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I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

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Prior Foreign Application(s)

Priority Claimed

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Full name of sole or first inventor (given name, family name): Jonathan D. Zook

Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_

Residence: Santa Clarita, California, United States of America

Citizenship: United States

Post Office Address: 20303 High Point Place  
Santa Clarita, California 91351☒ Additional inventors are being named on separately numbered sheets attached hereto.

Full name of second joint inventor, if any (given name, family name): Suzanna G. DeMoss

Second Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_

Residence: Van Nuys, California, United States of AmericaCitizenship: United StatesPost Office Address: 17420 Van Owen Street, #5  
Van Nuys, California 91406Full name of third joint inventor, if any (given name, family name): David W. Jordan

Third Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_

Residence: Northridge, California, United States of AmericaCitizenship: United StatesPost Office Address: 10960 Des Moines Avenue  
Northridge, California 91326Full name of fourth joint inventor, if any (given name, family name): Chandra B. Rao

Fourth Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_

Residence: Valencia, California, United States of AmericaCitizenship: United StatesPost Office Address: 25716 North Playa Dr, #T-15  
Valencia, California 91355Full name of fifth joint inventor, if any (given name, family name): Hakam Singh

Fifth Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_

Residence: Bradbury, California, United States of AmericaCitizenship: United StatesPost Office Address: 706 Winston Avenue  
Bradbury, California 91010Full name of sixth joint inventor, if any (given name, family name): Ahmed Sharaby

Sixth Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_

Residence: Canyon Country, California, United States of AmericaCitizenship: United StatesPost Office Address: 15866 Falconrim  
Canyon Country, California 91351

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